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THESIS

ANALYSIS OF COOLANT OPTIONS FOR ADVANCED METAL COOLED NUCLEAR REACTORS

by

Levent Can

December 2006

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It is well known that any neutron-producing device generates induced radioactivity as a by-product of its operation. In the case of nuclear reactors, the induced radioactivity includes fission and activation products. The overall focus of this study is the build up of induced radioactivity in the coolant of metal cooled reactors as well as the evaluation of other physical and chemical properties of such coolants. The objectives of the thesis are two fold. The first objective is to independently calculate the generation of Polonium-210 in reactors cooled by lead and lead-bismuth eutectic. The motivation for this is to address a noted lack of consensus among the world researchers on the significance of Po-210 build up in lead cooled reactors. The second objective is to evaluate the advantages and disadvantages of selected candidate metal coolants.

In addressing both objectives, the computer code ORIGEN was used. To establish the background basis for these assessments, fundamental concepts of reactor physics are reviewed and discussed.

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ANALYSIS OF COOLANT OPTIONS FOR ADVANCED METAL COOLED
NUCLEAR REACTORS

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requirements for the degree of

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ABSTRACT

It is well known that any neutron-producing device generates induced radioactivity as a by-product of its operation. In the case of nuclear reactors, the induced radioactivity includes fission and activation products. The overall focus of this study is the build up of induced radioactivity in the coolant of metal cooled reactors as well as the evaluation of other physical and chemical properties of such coolants. The objectives of the thesis are two fold. The first objective is to independently calculate the generation of Polonium-210 in reactors cooled by lead and lead-bismuth eutectic. The motivation for this is to address a noted lack of consensus among the world researchers on the significance of Po-210 build up in lead cooled reactors. The second objective is to evaluate the advantages and disadvantages of selected candidate metal coolants.

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LIST OF ACRONYMS

ANL	Argonne National Laboratory
BWR	Boiled Water Reactor
CANDU	Canada Deuterium Uranium Reactor
EAF	European Activation File
ENDF/B V	Evaluated Nuclear Data File/B-Version V
FENDL	Fusion Evaluated Nuclear Data Library
HLMC	Heavy Liquid Metal Coolant
LBE	Lead-Bismuth Eutectic
LMFBR	Liquid Metal Fast Breeder Reactor
ORIGEN	Oak Ridge Isotope Generator
ORIGENARP	Oak Ridge Isotope Generator Automated Rapid Processing
PWR	Pressurized Water Reactor
SSTAR	Small Secure Transportable Autonomous Reactor

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I.   INTRODUCTION

Energy demand in developed and developing countries is increasing dramatically as a result of growing populations. Meeting this enormous growth in demand for energy by the increased used of fossil fuels is becoming more and more expensive and environmentally damaging. On the other hand, renewable energy sources, such as hydro or geothermal, are far from being viable options due to limited supply and economic considerations. For these reasons, it is reasonable to expect that nuclear energy will re-emerge as a possible solution for growing energy demand in both developed and developing nations.

However, public acceptance of nuclear technology is an essential issue that complicates future development.[1] In order to be accepted by the general public, a new nuclear reactor concept must demonstrate efficient energy production, system safety, and the ability to operate with minimal waste material production.[2]

Promising new reactor technologies, together with new ideas for their deployment, are being discussed and explored all around the world.[3] Many studies in the United States, Japan, Korea, and Europe are underway to identify designs demonstrating inherent safety, competitive economics and satisfactory proliferation resistance. In that regard, several design concepts with alternative coolant technologies are being studied here in United States by major research laboratories such as Argonne National Laboratory, Los Alamos National Laboratory, and Lawrence Livermore National Laboratory. One of the many concepts currently being studied is the Small, Secure, Transportable, Autonomous Reactor, or SSTAR. In this thesis, the SSTAR reactor design has been used as the basis for the technical evaluations performed.

Before addressing the physics of activation in nuclear reactors, it is appropriate to present a background discussion of the SSTAR concept, summarize liquid metal coolant technology and identify the objectives of this thesis.

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[1] Galina Valeryevna, TSVETKOVA. "An Autonomous Long Term Fast Reactor System and The Principal Design Limitations of the Concepts" December 2003, Texas A&M University.

[2] Ibid.

[3] Ibid.

A.   SMALL SECURE TRANSPORTABLE AUTONOMOUS REACTOR (SSTAR)

The SSTAR concept is a fast reactor that would provide incremental energy generation to meet the needs of developing nations and remote communities lacking electrical grid connections, such as those that exist in Alaska or Hawaii, remote and coastal regions nations of the world.[4]

SSTAR offers many attractive features over conventional reactors such as transportability, simplification of operation, competitive market performance, and nonproliferation characteristics. [5]
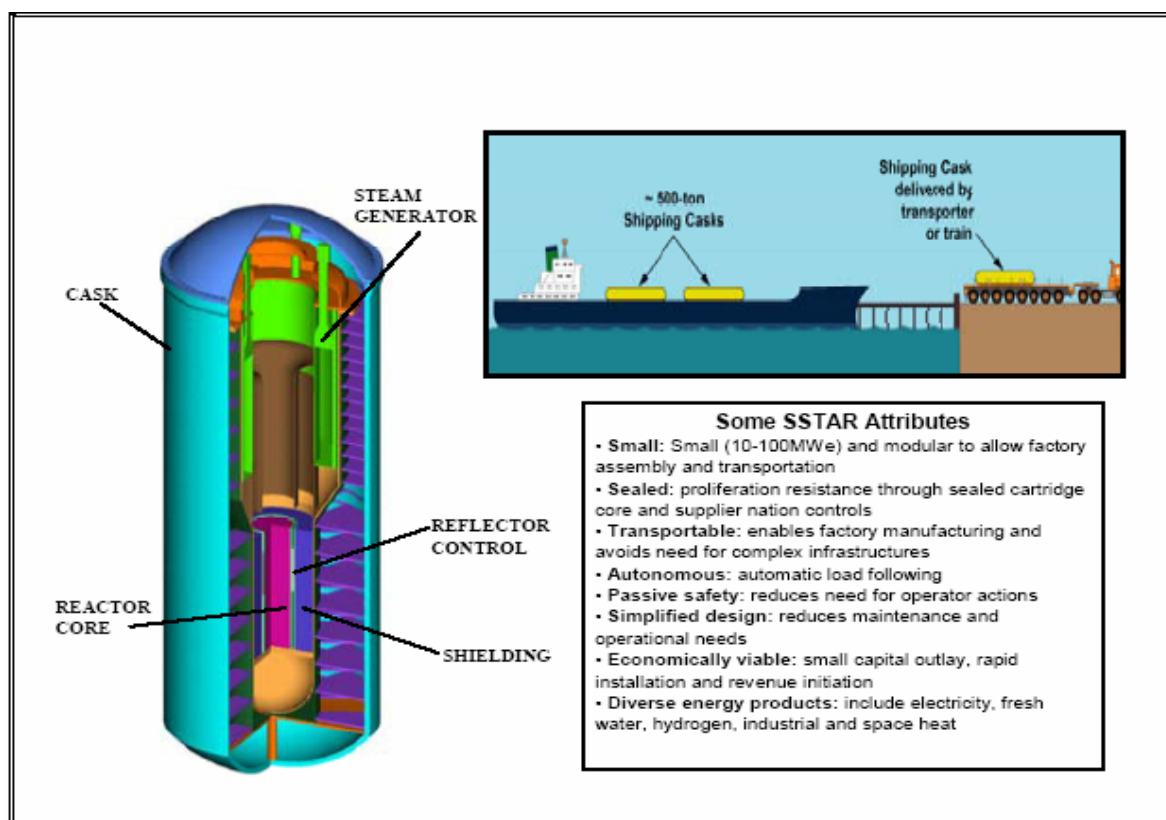
Figure 1.1   The Small Secure Transportable Autonomous Reactor (SSTAR)[6]

[4] Gabrielle, Rennie, "Nuclear Energy to Go; A Self-Contained Portable Reactor" Lawrence Livermore Science and Technology Review, August 2004.

[5] Ibid.

[6] Ibid.

1. **Transportability**

SSTAR is designed to be factory fabricated, transported, and delivered in shipping casks. SSTAR will be about 15 meters tall and 3 meters wide and will not weigh more than 500 tons. Since the supplier nation will provide fuel cycle services, the host nation will be able to produce electricity or other energy products without needing an independent supply of fuel or fuel services. Moreover, the host nation will not need to deal with spent fuel or nuclear waste at the end of the cycle, thereby avoiding substantial financial and environmental burdens.

2. **Simplification of Operation**

The operating and maintenance costs of conventional power reactors are substantial. SSTAR is a design based on the principle of fully automatic simple control. If any operational abnormality is detected, the reactor will be switched off automatically. Ultimately, the operation staff will have little or no tasks when the reactors are running normally. Maintenance and inspection of a reactor could be performed as completely independent operations.

Also, conventional reactors require a small army of experts to operate and monitor the reactor systems. Unlike conventional reactors, SSTAR will require minimal personnel, maintenance, and inspection of reactor components. They will rely on simplicity in operation with reliance on autonomous control and remote monitoring

3. **Market Competition**

In the nuclear power industry, relatively few innovations have been made over past decade. Unsurprisingly, the main reason for this is that there has been little market competition among manufacturers in the area of research and development of large-scale systems. It is likely that SSTAR, with its promising place in the future market, will open the door for new innovation and further improvements in the safety and reliability of nuclear power systems in the decades to come.

Additionally, SSTAR will be radically different from existing nuclear plants. When the nuclear industry reaches the point where large numbers of small nuclear power

generation units can be manufactured, it will be possible to reduce the unit cost per kilowatt (kW) of these reactors to less than one-third of the present level[7]

4. Non-proliferation

The core of SSTAR is a single cassette or an integrated, non removable part of the reactor vessel. Access to fuel or neutrons from the core is restricted during the 30-year burn cycle. Therefore there is essentially no chance to divert fuel at the host. Secondly, the plant design does not include either refueling equipment or provisions for onsite fuel storage. At the end of the 30-year lifetime, SSTAR will be shipped back to the supplier country and replaced if one is still needed. Since the core is still big enough to require substantial effort to move, any unauthorized attempt to replace or remove the core is easy to monitor or detect by satellite or any other remote surveillance methods.

B. LIQUID METAL COOLANT TECHNOLOGY

Regardless of the design of a nuclear reactor, heat removal from the core is one of the fundamental problems to be addressed. Heat removal is determined by the energy generation in the core, the characteristics of the reactor, and the coolant material.

Various coolant media have been studied and used around the world. The idea of a liquid metal-cooled fast reactor goes back to the very earliest days of atomic energy, beginning even with one of the very first plants in the U.S., the experimental breeder reactor (EBR-I), which began operation in 1951.[8]

The most highly studied and extensively used liquid metal coolant, due to its superior heat transfer properties, is sodium. Hence, the effort to develop sodium technology during the past 40 years has been enormous and has led to the situation today wherein sodium cooling for fast reactors is considered to be a proven technology.

Like sodium four decades ago, lead and lead bismuth coolant technology have gained increased attention among nuclear reactor designers. There are several reasons for the increasing interest in lead and lead-bismuth eutectic (LBE)[9] coolant. First, the

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[7] Hattori, Sadao, "Technical potential of small reactors" January, 2004, Tokyo, JAPAN

[8] Bruce W. Spencer, "The Rush to the Heavy Metal Reactor Coolants-Gimmick or Reasoned", Proceedings of ICONE 8, April 2-6, 2000, Baltimore, MD, USA.

[9] Lead Bismuth Eutectic (LBE) , also sometimes called "liquid lead", is an alloy mixture consisting of 45.5 wt% Lead and 54 5wt% Bismuth. As an alloy, LBE has a melting temperature of $123.5^{o}C$ in contrast to the $327^{o}C$ melting temperature of lead.

transmutation of nuclear waste in sub-critical, accelerator-driven reactors has been studied in several countries, including the U.S., introducing a potential new mission for fast reactors; consideration of lead or lead alloy as the target materials for neutron spallation has prompted their simultaneous consideration as a reactor coolant.[10] Secondly, the new openness in the nuclear sector of Russia has brought us new information about their extensive technology development of LBE coolants used in reactors for submarine propulsion (Gromov, 1999), and has additionally introduced us to their development plans for new lead-cooled fast reactors (Adamov, 1997).[11] Moreover, the U.S. Department of Energy's recent Nuclear Energy Research Initiative (NERI) has encouraged ideas for new and innovative technologies supporting the nuclear option in the 21st century.[12]

Unlike other metal coolants, tin has not been fully explored or used as a reactor coolant. One of the first proposals to use liquid tin for nuclear power facility was done by J. R. Weeks in 1971.[13]. He considered melts of natural tin and its eutectic alloys, Bi-Sn, Pb-Sn, and Bi-Pb-Sn as potential coolants for fast reactors

Due to the fact that GEN-IV[14] systems include fast spectrum reactors for efficient material management, it is appropriate to consider usage of coolants with low moderating power such as sodium, lead, LBE, and tin. Hence, LBE and tin, in addition to the proposed GEN-IV coolants, lead and sodium, are studied in this thesis.

C.   OBJECTIVES OF THE THESIS

It is well known that any neutron-producing device generates induced radioactivity as a by-product of its operation. In the case of nuclear reactors, the induced radioactivity includes fission and activation products. The overall focus of this study is the build up of induced radioactivity in the coolant of metal cooled reactors as well as the

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10 Bruce W. Spencer, "The Rush to the Heavy Metal Reactor Coolants - Gimmick or Reasoned", Proceedings of ICONE 8, April 2-6, 2000, Baltimore, MD, USA.

11 Ibid.

12 Ibid.

13 J. R. Weeks, "Lead, Bismuth, Tin and Their Alloys as Nuclear Coolant", Nuclear Engineering and Design, 1971.

14 Generation IV nuclear energy systems are future, next-generation technologies that will compete in all markets with the most cost-effective technologies expected to be available over the next three decades. http://nuclear.inl.gov/gen4. Accessed 18 October 2006, The United Sates Department of Energy.

evaluation of other physical and chemical properties of such coolants. The objectives of thesis are two fold. The first objective is to independently calculate the generation of Polonium-210 in reactors cooled by lead and lead-bismuth eutectic. The motivation for this is to address a noted lack of consensus among the world researchers on the significance of Polonium-210 build up in metal cooled reactor. The second objective is to evaluate the advantages and disadvantages of selected candidate metal coolants.

In addressing both objectives, the computer code ORIGEN was used. To establish the background basis for these assessments, fundamental concepts of reactor physics are reviewed and discussed. In addition to these objectives, the general goals of this study are stated as follows:

> Understand and apply the theoretical numerical concepts underlying analytical calculations of coolant activation, such as a solution to the Bateman equations.
>
> Modify and run the computer code ORIGEN according to our needs and apply ORIGEN-S as a computational tool to calculate activation and decay within the metal coolants:
>
> - o Analyze and compare Po-210 build up in lead and lead bismuth-cooled systems.
> - o Analyze gamma radiation from Na-24 activation in sodium and compare the results with previous work.
> - o Understand the activation processes in tin coolants.
>
> Revisit the fundamental concepts of reactor physics that are embedded in computer simulations like ORIGEN.
>
> Understand the physical, chemical and thermal properties and corrosive behavior of "selected candidate coolants"[15], and perform comparative assessment of advantages and disadvantages.

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[15] In this study, by using "selected coolants", I refer to coolants studied in this thesis: lead, LBE, sodium, and tin.

D.   ORGANIZATION OF THE THESIS

The thesis is composed of five chapters. Chapter I provides an introduction an overview of the SSTAR concept, a discussion of liquid metal coolant technology and a statement of objectives.

Chapter II presents the fundamental concepts of reactor physics, which are used in the analyses of the thesis. It touches on cross sections of selected coolants, the phenomenon of radioactive decay (parent-daughter decay schemes and the Bateman equations), coolant activation, the four-factor formula, and criticality.

Chapter III discusses physical and thermal properties and the corrosive behavior of selected liquid metal coolants.

Chapter IV explains how ORIGEN, the computational code used in this thesis, solves the Bateman equations and discusses the calculation of activation and fission product generation in selected coolants and reactor systems. The results also are compared to those available elsewhere in the literature in order to test the findings.

Chapter V presents the conclusions of the study. In this chapter, the advantages and disadvantages of selected coolants are identified. Furthermore, recommendations are made for future thesis work in this area.

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II.   THE PHYSICS OF FAST REACTORS

Although our knowledge about nuclear technology has increased substantially over the years, the basic physics that governs reactor design has hardly changed. Given the fact that SSTAR, a small fast reactor, will be based on these same fundamental principles, I will briefly summarize the physics of fast reactors. A more detailed treatment of nuclear reactor theory would be the subject of an entire thesis study by itself. However, the intent of this chapter is to recall the fundamental concepts of reactor physics that are embedded in computer simulations like ORIGEN. It touches on cross sections of selected coolants, parent-daughter decay, the Bateman equations, coolant activation, the four-factor formula, and criticality.

A.   CROSS SECTIONS

For a particular nuclear reaction at the atomic level, each atom can be imagined to have a sphere of influence associated with it such that, if a neutron penetrates the sphere, an interaction takes place. This sphere presents the same *cross-sectional area* in all directions and is called the *cross section* of the atom. Neutron cross sections are expressed in units of barns, where 1 barn is equal to $10^{-24} cm^2$.

The probability of interaction with a neutron to form a radioactive isotope is referred to as neutron activation cross section. Thermal activation cross sections of the principal isotopes associated with candidate coolants are given in the tables below; energy dependent cross-sections are presented in the Figures 2.1 to 2.4.[16]

1.   Lead (Pb)

Nuclide	Abundance (%)	Thermal Activation Cross Sections	Reaction	Radioactive nuclide	Half life
Pb-208	52.3	0.6 ± 0.2mb	(n, γ)	Pb-209	3.3 hrs

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[16] Note that in calculating neutron activation in coolants, an effective cross section must be used. This effective or average cross section takes in to account the energy dependent cross section relation as well as the energy dependency of the neutron spectrum.
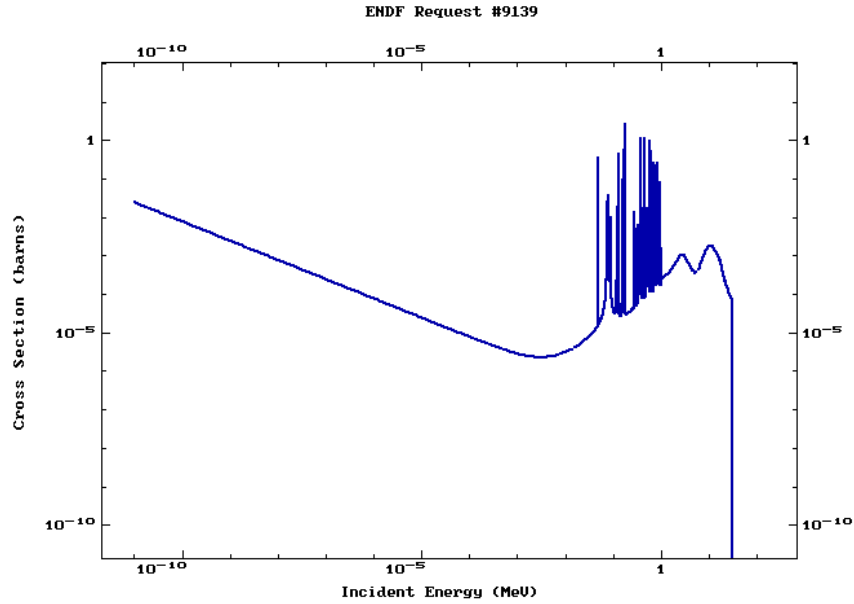
Figure 2.1   Cross Section for Pb^{208}, (n, γ) Reaction

(ENDF/6 Using ENDFPLOT from Brookhaven National Laboratory over Internet)

2.   Lead-Bismuth Eutectic (LBE)[17]

Nuclide	Abundance (%)	Thermal Activation Cross Sections	Reaction	Radioactive nuclide	Half life
Bi-209	100	19 ± 2mb	(n, γ)	Bi-210	5 days
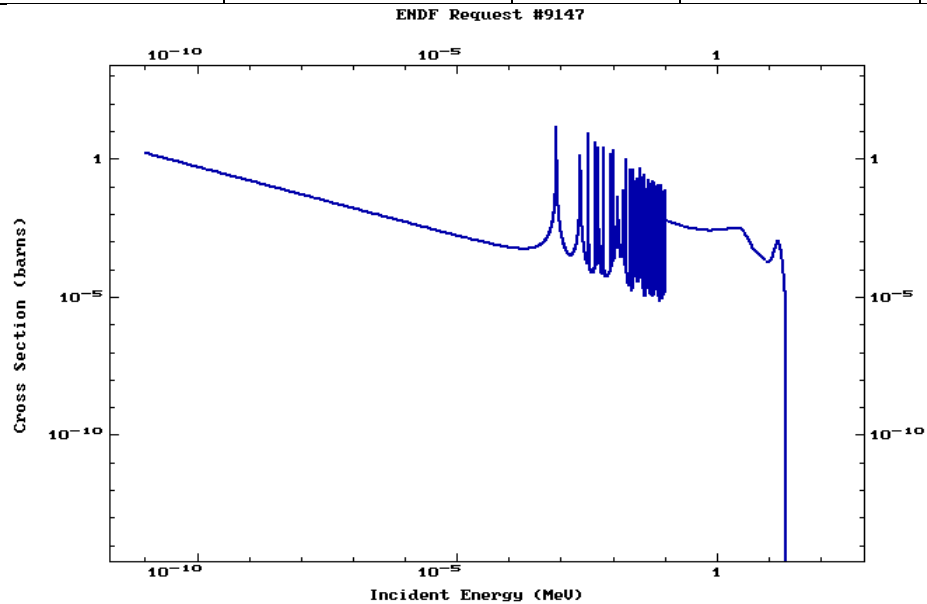
Figure 2.2   Cross Section for Bi-209, (n, γ) Reaction

(ENDF/6 Using ENDFPLOT from Brookhaven National Laboratory over Internet)

[17] LBE consists of lead and bismuth. The activation of Bi-209 results in generation of Bi-210 which rapidly decays to Po-210 with a half life of 138.4 days.

3.   Sodium (Na)

Nuclide	Abundance (%)	Thermal Activation Cross Sections	Reaction	Radioactive nuclide	Half life
Na-23	100	0.56 ± 0.03mb	(n, γ)	Na-24	15 hrs
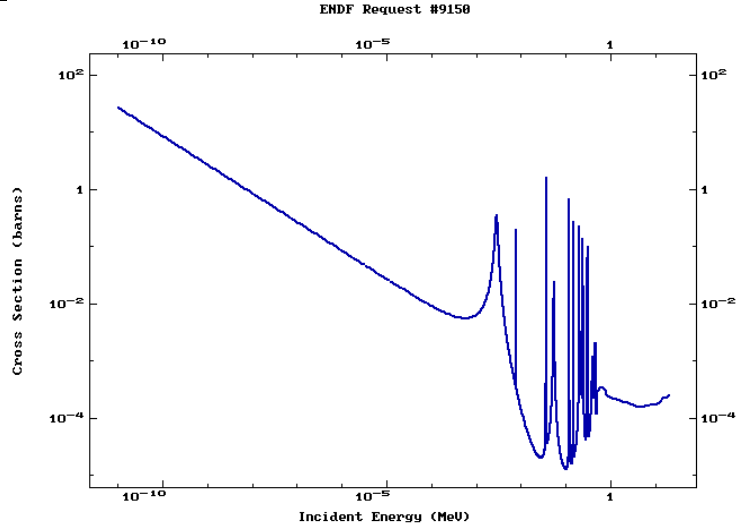
Figure 2.3   Cross Section for Na-23, (n, γ) Reaction
(ENDF/6 Using ENDFPLOT from Brookhaven National Laboratory over Internet)

4.   Tin (Sn)

Nuclide	Abundance (%)	Thermal Activation Cross Sections	Reaction	Radioactive nuclide	Half life
Sn-120	33	1 ± 1mb	(n, γ)	Sn-121	>400 days
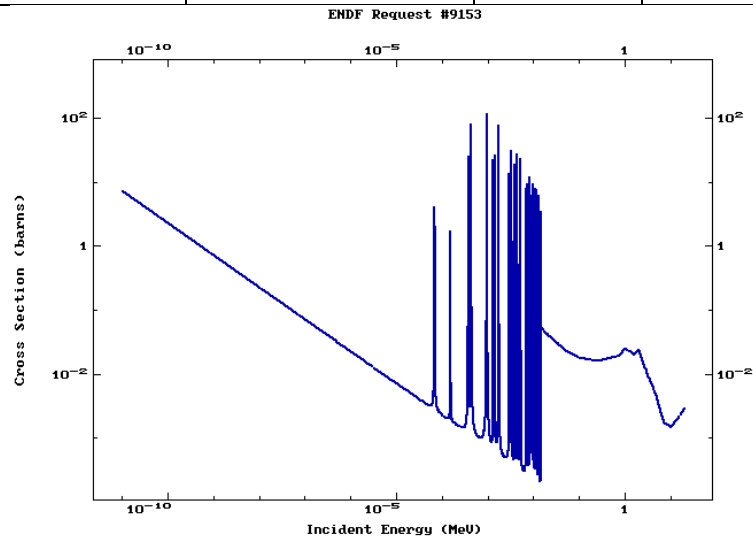
Figure 2.4   Cross Section for Sn-120, (n, γ) Reaction
(ENDF/6 Using ENDFPLOT from Brookhaven National Laboratory over Internet)

B. RADIOISOTOPE PRODUCTION AND THE BATEMAN EQUATION

There are three general ways of producing radioisotopes: fission of heavy atoms, activation by particles and the decay of radioactive series. Each method can be discussed in terms of a first order differential equation that governs the quantity of a given isotope at any given time $N(t)$.

The solution to the first order differential equation that governs the production and decay of a radionuclide will provide fundamental understanding for the activation phenomenon.

If we assume that Q(t) is the constant rate of production of a radioisotope for a time interval (0,T), then production will take place at constant Q_0 over time as shown below.
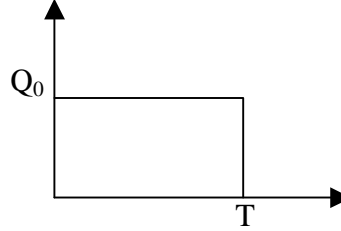
Figure 2.5   Constant Production Rate over Time Interval (0,T)

Considering both production and decay during the interval (0,T), and recognizing that radioactive decay occurs in accordance with a decay constant λ, the equation governing $N(t)$ is going to be:

$$\frac{dN(t)}{dt} = Q_0 - \lambda N(t) \text{, where } \lambda \text{ is the decay constant.} \tag{2.1}$$

Then the solution to equation (2.1) above, with the initial condition that there is no radioisotope prior to production, N(t=0)=0, is;

$$N(t) = \frac{Q_0}{\lambda}(1 - e^{-\lambda t}) \text{, where t<T} \tag{2.2}$$

Where for t>T, the solution to the differential equation in (2.1) without the source term is going to be;

$$N(t) = \frac{Q_0}{\lambda}(1 - e^{-\lambda T})(e^{-\lambda(t-T)}) \tag{2.3}$$

12

Hence, radioisotope production over time due to coolant activation, will reflect increase of N(t) during production, trending toward an asymptotic value of Q_0 / λ, and the subsequent exponential decay after production is stopped at t=T. Figure 2.6 demonstrates the buildup and decay of a radioisotope over time.[18]
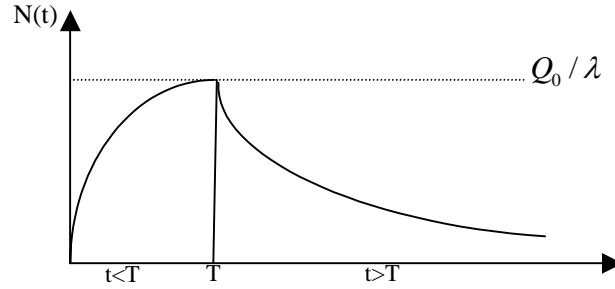
Figure 2.6   Radioisotope Build up and Decay over Time

Additionally, radioisotopes can also be produced as the decay products of parent nuclides in a series of decays. I assume a case in which an initial quantity of radionuclide, N_1 decays to a daughter product, N_2, that is also radioactive, and which subsequently decays to a stable end product, N_3.

$$N_1 \rightarrow N_2 \rightarrow N_3 (Stable)$$
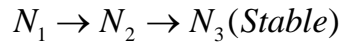
The differential equations and time domain solutions for this system of three nuclides assuming no external generations are as follows:

$$\frac{dN_1(t)}{dt} = -\lambda_1 N_1(t) \tag{2.4}$$

$$\frac{dN_2(t)}{dt} = \lambda_1 N_1(t) - \lambda_2 N_2(t) \tag{2.5}$$

A solution to the set of differential equations can be accomplished by Laplace transforms and by manipulating equations (2.4) and (2.5). The final solution for the time-dependent amount of N_2 yields:

$$N_2(t) = N_2(0)e^{-\lambda_2 t} + \frac{N_1(0)\lambda_1}{\lambda_2 - \lambda_1}\left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right] \tag{2.6}[19]$$

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[18] W.E, Meyerhof, "Elements of Nuclear Physics", McGraw-Hill, New York, 1967.

[19] http://www.eas.asu.edu/~holbert/eee460/RadioactiveDecay.pdf Accessed 12 November 2006, Arizona State University Electrical Engineering Department. EEE460 Lecture notes.

The differential equation for the end product (granddaughter) N_3 consists only of the decay the parent material since there are no losses because the end product is stable:

$$\frac{dN_3(t)}{dt} = \lambda_2 N_2(t) \qquad (2.7)$$

A solution to equation (2.7) can be found by integrating this expression and substituting into equation (2.6). The final expression of time-dependent N_3 is as follows:

$$N_3(t) = N_3(0) + \int_0^t \lambda_2 N_2(\tau) d\tau \qquad (2.8)$$

$$= N_3(0) + N_2(0)(1 - e^{-\lambda_2 t}) + N_1(0)\left[1 + \frac{\lambda_2}{\lambda_1 - \lambda_2} e^{-\lambda_1 t} - \frac{\lambda_1}{\lambda_1 - \lambda_2} e^{-\lambda_2 t}\right] \qquad (2.9)[20]$$

Equations (2.4) through (2.9) are known as *the Bateman equations*. They are used to analyze situations when the decay constants λ_1 and λ_2 take different values. For example, where $\lambda_1 \gg \lambda_2$, the parent is called short-lived and in the opposite case, where $\lambda_2 \gg \lambda_1$, the parent is called long-lived.

C.   COOLANT ACTIVATION & DECAY

Radioactivity is measured in terms of the rate of radioactive decay. The quantity $\lambda N(t)$, where N is the number of radioisotope atoms at time t, is called *activity*. The standard units of radioactivity have been *Curie* and *Becquerel*. The values of the Curie and the Becquerel are given below:

1 Ci= 3.7 X 10^{10} Bq and   1 Bq= one disintegration per second..

The coolant in reactor systems provides for the removal of heat produced at a desired power level, ensuring that maximum temperature is always maintained lower than the predetermined limit value. The heat, *q*, released by the core, is absorbed by the coolant, which passes through the core entering at time (t_{in}) and exiting at time (t_{out}). A schematic expression of coolant flow through the core is as follows:

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[20] http://www.eas.asu.edu/~holbert/eee460/RadioactiveDecay.pdf Accessed 12 November 2006, Arizona State University Electrical Engineering Department. EEE460 Lecture notes.
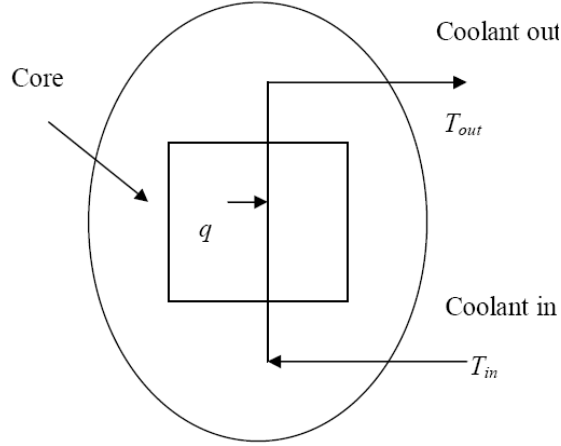
Figure 2.7   Coolant Flow through a Reactor

Due to circulation, target atoms in the coolant are exposed to high neutron flux as they pass through the core. The circulating coolant is activated as it passes through the core and decays in the external portions of the coolant system as well as in the core.

In order to understand coolant activation further, I assume that the number of radioactive atoms formed in a unit volume of coolant as a result of the first pass through the core is represented by K. Where the neutron flux, $\Phi = n\upsilon$, is constant and the time spent in the reactor is t_1 ($t_1 = t_{in} - t_{out}$), then K yields

$$K = \frac{Q}{\lambda}(1 - e^{-\lambda t_1}) \tag{2.10}$$

where Q=$n\upsilon\, N_0 \sigma$ and $n\upsilon$ is the flux, N_0 is the original number of atoms of target isotope and σ is the cross section for the same atom.

As the unit volume of coolant passes through the external circuit with time t_2 ($t_2 = t_{out} - t_{in}$), the activity decays by a factor $e^{-\lambda t_2}$, and the number of radioactive atoms after one complete cycle is:

$$A_1 = Ke^{-\lambda t_2} \tag{2.11}$$

The coolant activity for the first cycle is going to be as defined in equation (2.11). In the second cycle, the number of radioactive atoms formed is again $Ke^{-\lambda t_2}$, but the

number created in the first cycle decays by a factor of $e^{-\lambda(t_1+t_2)}$.[21] After n times passing through the core, coolant activity at entry and exit is going to be as follows:

Coolant activity at exit from core:

$$A_n(exit) = K\left[\frac{1-e^{-n\lambda(t_1+t_2)}}{1-e^{-\lambda(t_1+t_2)}}\right]$$

(2.12)

Coolant activity at entry to core:

$$A_n(entry) = Ke^{(-\lambda t_2)}\left[\frac{1-e^{-n\lambda(t_1+t_2)}}{1-e^{-\lambda(t_1+t_2)}}\right]$$

(2.13)

If the full cycle time (t_1+t_2) is small compared to mean life $1/\lambda$, then equations (2.12) and (2.13) for coolant activation are going to be as follows:

$$A_n(exit) = \frac{K}{\lambda(t_1+t_2)}\left[1-e^{-n\lambda(t_1+t_2)}\right]$$

(2.14)[22]

$$A_n(exit) = \frac{Ke^{-\lambda t_2}}{\lambda(t_1+t_2)}\left[1-e^{-n\lambda(t_1+t_2)}\right]$$

(2.15)[23]

Besides the mathematical derivation of coolant activation, the neutronic characteristics of coolant in reactor design are significant. Coolant activation must be taken into consideration for safety and reliability reasons. Activation in a radiation environment results in radioactive nuclides circulating through the coolant loop. Those nuclides that have short half-lives are plant radiological hazards, which introduce requirements and equipment for operations, repairs, and maintenance.

D.   THE FOUR-FACTOR FORMULA

A nuclear reactor is built on the principle that fission reactions induced by the neutrons can create and maintain a steady chain reaction. Technically, each fission outcome neutron will either induce another fission event; leak out; or be absorbed by fissile and non-fissile material. For a critical system, there is a balance in which each fission generates enough neutrons to provide for a successor fission after taking into account leakage and absorbtion.

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[21] Harold, Etherington, " Nuclear Engineering Handbook", McGraw-Hill, New York ,1958,p. 7-15

[22] Ibid.

[23] Ibid.

However, there are fundamental differences between processes in thermal and fast reactors. The principal distinction between the thermal and fast reactor is that whereas, in thermal reactors, moderating material is deliberately introduced in order to slow neutrons down as rapidly as possible to avoid resonance capture, in fast reactors there is no moderating material. Thus, fast reactors are constructed without a moderator so that the average neutron energy is much higher than in a thermal reactor.

The four factor formula was developed as a rule of thumb to consider criticality in thermal reactors. Although the four factor formula does not hold for fast reactors, it can help us to understand the concepts behind criticality which are also valid for fast reactors. With this in mind, the conditions to sustain criticality (i.e., k=1), in a thermal reactor, can be related to four factors.

The multiplication factor, k, defines the relationship between the neutrons generated between two successive generations and enables operators to monitor neutron cycles during operation.
In equation form, this is:

$$k = \frac{\text{Number of fissions in one generation}}{\text{Number of fissions in the preceding generation}} \qquad (2.16)$$

In order to facilitate understanding of the neutron cycle in a reactor, a simple schematic expression is presented below:
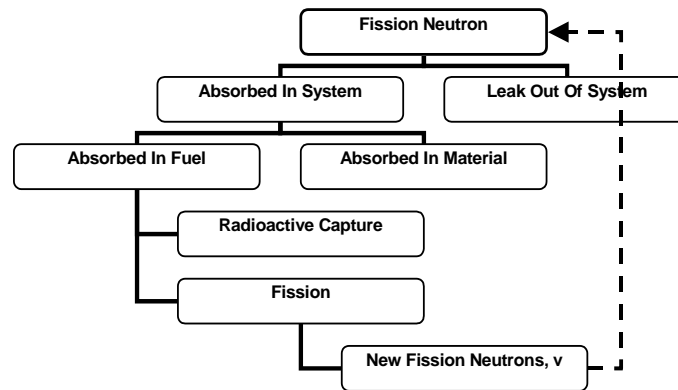
Figure 2.8   Neutron Cycle in a Reactor

With the definition of k in mind, the four factor formula considers the phenomena of thermal utilization (f), regeneration (η), fast fission (ε) and resonance escape (p).

17

1. Thermal Utilization (f)

Thermal utilization is defined as the ratio of the number of neutrons absorbed per unit time in fuel to the total number of neutrons absorbed per unit time in the reactor. For a homogenous reactor, where structural materials and coolant are homogeneously mixed, it is given as:

$$f = \frac{\text{Thermal neutrons absorbed by fuel}}{\text{Total thermal neutrons absorbed}} = \frac{\sum_a^{Fuel}}{\sum_a^{Total}} = \frac{\sum_a^{Fuel}}{\sum_a^{Fuel} + \sum_a^{Coolant} + \sum_a^{Structure}} \quad (2.17)$$

where $\sum_a$ is a macroscopic abortion cross section.

2. Regeneration Factor (η)

The quantity of η is defined as the number of fission neutrons produced per neutron absorbed in fuel.

$$\eta = \frac{\text{Number of neutrons produced by fission}}{\text{Number of neutrons absorbed by fuel}} = \frac{\sum \upsilon N \sigma_f}{\sum N \sigma_a}, \quad (2.18)$$

where
  υ is the number of neutrons emitted per fission,
  N is the number of atoms
  σ_f is the *microscopic fission* cross section
  σ_a is the *microscopic absorption* cross section

3. Fast Fission Effect (ε)

In thermal reactors containing a large amount of fissionable but not fissile material, interactions with non-fissile nuclides induce a small fraction of fissions. These fast fission events can be accounted for by the factor:

$$\varepsilon = \frac{\text{Total fission neutrons from thermal and fast fission}}{\text{Fission neutrons from thermal fission}} \quad (2.19)$$

4. Resonance Escape Probability (p)

After increasing release from fissions at high energy, neutrons begin to diffuse through the reactor. As the neutrons move, they collide with nuclei of fuel moderator and non-fuel material in the reactor, losing part of their energy in each collision and slowing down. While they are slowing down through the resonance region of Uranium-238, there is a chance that some neutrons will be captured. The probability that a neutron

will not be absorbed by a resonance peak is called the resonance escape probability, defined below:

$$p = \frac{\text{Number of neutrons slowing to thermal energy}}{\text{Total number of fast neutrons available for slowing}} \qquad (2.20)$$

Finally, the multiplication factor, k, can be broken down into a product of the four-factor formula described above:

$$k = \eta \varepsilon p f \qquad (2.21)$$

If k is greater than 1, the number of fissions increases generation to generation. In this case, the energy released by the chain reaction increases over time, and the chain reaction, or the system in which it is taking place, is said to be *supercritical*.[24] However, if k is less than 1, the number of fissions decreases with time and the chain reaction is called *subcritical*. Where k is equal to 1, the chain reaction proceeds at a constant rate, energy is released at a steady level, and the system is said to be *critical*.[25]

Basically, nuclear reactors are assemblies of materials and equipment in which chain reactions are controlled. In a reactor, this control is accomplished by an operator or an automatic control. In order to increase the power being produced by a reactor, the operator increases k to a value greater than unity so that the reactor becomes slightly *supercritical* for a limited period of time. Reduction of power is the opposite process.
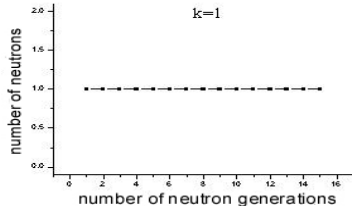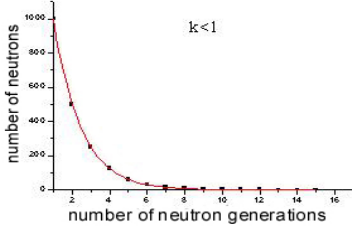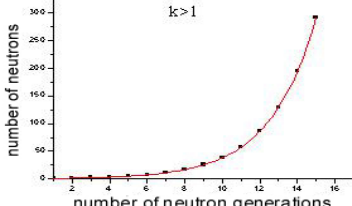
Operators adjust the value of k when they want to reduce/increase power or shut down the reactor. As a result, the power output of the system decreases or increases.

Criticality is a fundamental concept in nuclear reactors. There are three different possible criticality conditions. Table 2.1, in the next page, explains criticality:

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[24]John R. Lamarash, and Anthony J. Baratta, "Introduction to Nuclear Engineering", 2001, 117-118.

[25] Ibid.

Criticality	Corresponding Explanation	Graphical Representation
Critical	The sum of the neutron absorption and leakage rates is **exactly equal** to the neutron production rate	
Sub-critical	The sum of the neutron absorption and leakage rates is **higher** than the neutron production rate	
Supercritical	The sum of the neutron absorption and leakage rates is **smaller** than the neutron production rate	

Table 2.1   Criticality Conditions in Reactors

III. THERMOPHYSICAL AND THERMOHYDRAULIC PROPERTIES OF COOLANTS

The appropriate choice of coolant for reactor systems is a very important factor to gain high performance. Moreover, the properties of a reactor coolant are one of the fundamental determinants of reactor design and, to a certain extent, have a significant impact on the technical and economic characteristics of power plants.

Finding the right cooling medium has been a daunting task for nuclear engineers for half a century. There is no single dominant property that leads to conventional wisdom. Generation IV (GEN-IV), the international program to consider advanced nuclear reactor technologies, involves assessment of a broad variety of reactor coolants. However, there is no "silver bullet" answer to the question of which coolant is going to best meet the requirements put forward by Generation IV.

Although there are other liquid metals, such as gallium and lithium, that could be considered as reactor coolants, in this thesis I include assessment of lead-bismuth eutectic (LBE) and tin in addition to two of the GEN-IV[26] coolants: lead and sodium.

In this chapter the properties, compatibility and economics of the selected candidate coolants are addressed.

A. PROPERTIES OF COOLANTS

The extraordinary thermophysical properties of liquid metals make them favorable as reactor coolants. In particular, heat transfer properties are the most noticeable. Due to their high thermal conductivity, they have been considered favorably by nuclear engineers as heat transfer media for nuclear reactors.

1. Physical Properties

The physical and nuclear properties of bismuth, lead, LBE, tin, and sodium are given in Table 3.1. As seen in the table, the attractiveness of these materials comes from their low melting points, high heat transport capabilities, and high boiling points in general.

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[26] The U.S. Department of Energy's Office of Nuclear Energy, Science and Technology has engaged governments, industry, and the research community worldwide in a wide ranging discussion on the development of next generation nuclear energy systems known as "Generation IV".

Properties	Na	Pb	Bi	LBE	Sn
Atomic number	11	82	83	--	50
Atomic Mass	22.99	207.2	208.98	--	118.7
Melting Point (^{0}C)	98	327.4	271.41	123.5	232
Boiling Point ,(^{0}C)	883	1745	1552	1670	2270
Density, kg/m^3	970	10700	9800	10200	7000
Heat Capacity, (kJ/kg.Ḱ) at 450 ^{o}C (liq)	1.269	147.3	150	146	228
Thermal Conductivity (W/mḰ) at 450°C(liq)	68.8	17.1	14.2	14.2	66.6
Kinematic Viscosity, (m^2/s) at 450°C	3×10^{-7}	1.9×10^{-7}	1.3×10^{-7}	1.4×10^{-7}	1.38×10^{-7}

Table 3.1   Thermophysical Properties of Liquid Metals[27]

Sodium is the material that has been traditionally considered for use as the primary coolant in fast reactors.[28] Compared to the properties of other metals, sodium has a lower melting point and relatively higher thermal conductivity. According to a Generation IV Nuclear Energy Systems report[29] released February 21, 2006, the core outlet temperature and peak cladding temperature will be 561°C and 650°C, respectively. Hence, for higher operating temperatures, the boiling temperature of sodium might be critical.

In fast nuclear reactors, both because of the possibility of increased reactivity due to boiling and the potential loss of effectiveness of coolant heat transfer, coolant boiling must be avoided. Therefore, due to their high boiling points, lead, LBE or tin might be considered favorably for SSTAR type systems with higher operating temperatures.

Lead, compared to the other selected coolants, has a relatively high melting point. For this reason an auxiliary heating system might be required to ensure a liquid phase of coolant before startup.

[27] Harold, Etherington, "Nuclear Engineering Handbook", McGraw, New York, 1958, p. 9-17 to 24; IAEA-1289,p.20.

[28] A. E. Waltar; A. B. Reynolds, "Fast Breeder Reactors", Pergamon Press, New York, (1981),p.436.

[29] http://www.ne.doe.gov/nerac/FinalRoadmapforNERACReview.pdf Accessed 09 September 2006, The United States Department of Energy.

The chief advantage of LBE over lead is its considerably lower melting point, which greatly simplifies the design and operation of whole plant because the likelihood of coolant freezing is reduced.[30] The variation of the melting point of lead-bismuth alloys as a function of the relative fraction of lead-bismuth is best illustrated by means of the liquidus curve in Figure 3.1.
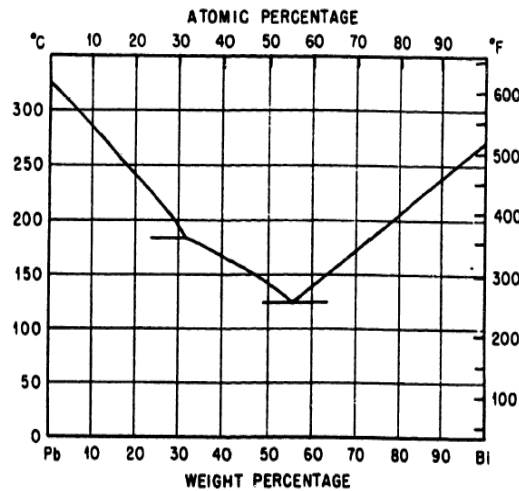
Figure 3.1   Liquidus Form of the Lead-Bismuth Alloy (Taken from Lyon, 1952)

The melting point of lead-bismuth, which, used in its eutectic form is composed of 44.5 wt% lead and 54.5 wt% bismuth, is considerably lower than that of either lead or bismuth. This would make a LBE cooled reactor simpler in terms of operations and performance.

According to Table 3.1, the density of lead, LBE, and tin is almost ten times higher than that of sodium. The high density of lead, LBE, and tin has some drawbacks for the design of the overall reactor system. It complicates the seismic design of the pant and may require additional pumps or higher pumping power for coolant circulation.[31] The low pumping power is one of the main contributing factors that led to the selection of

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[30] Buongiorno, Jacopo, "Conceptual Design Of A Lead-Bismuth Cooled Fast Reactor within-Vessel Direct-Contact Steam Generation" 2001, MIT.

[31] Nuclear Energy Agency, "Accelerator Driven Systems and Fast Reactors in Advanced Nuclear Fuel Cycles".

sodium for Liquid Metal Fast Breeder Reactor (LMFBR) systems worldwide.[32] The pumping power requirements for lead, LBE and tin are much higher than for sodium.

With regard to the pumping power argument, lead, tin, and LBE have different and groundbreaking promises. According to thorough heat transport analysis (Spencer, 2000), selected coolants have excellent natural coolant and heat removal characteristics in low-pressure drop-core design. But coolants whose neutronic characteristics permit a large coolant fraction in the core (i.e., bismuth, lead, and LBE) may be considered for 100% natural-circulation heat transport, introducing possibilities for a new level of plant simplification and inherent safety.[33]

High thermal conductivity is another desired property in reactor coolants. The overall efficiency of the reactor system is affected to a significant degree by the thermal conductivity of the coolant.[34] The thermal conductivities of coolants at different temperatures are given in Table 3.2.

T(^{o}C)	Na W/(mC)	Lead W/(mC)	LBE W/(mC)	Tin W/(mC)
200	81.02	-	11.7	-
300	77.12	-	12.7	33.9
400	73.19	17.66	13.7	33.1
500	69.28	18.79	14.7	32.4
600	65.35	19.82	15.5	31.8
700	61.48	20.94	16.4	31,0

Table 3.2   Thermal Conductivity of Selected Liquid Metals[35]

As seen from the table above, the thermal conductivity of sodium and tin provides a considerable advantage over lead and LBE. This is one of the important properties of tin, along with its high boiling point, that led nuclear scientists to consider tin as a possible candidate for nuclear reactors.

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[32] Bruce W. Spencer, "The Rush to Heavy Liquid Metal Reactor Coolants Gimmick or Reasoned", ICONE-8729, April 2-6, 2000, Baltimore, MD USA.

[33] Ibid.

[34] Kopelman, Bernard, "Materials for Nuclear Reactors" 1959 p. 314.

[35] Compiled from IAEA-TECDOC-1289, p 22.

Tin has a very high boiling temperature and a lower melting temperature than both lead and bismuth. This advantage can, of course, be utilized only in very high-temperature reactors, a field in which development is limited.[36]

In summary, the thermophysical properties of coolants represent only some of many criteria that can be considered in comparative assessment. Clearly, sodium has been chosen by many nations as a primary fast reactor coolant due to its superior thermophysical properties, such as low density, lower melting point, and considerably higher thermal conductivity. Lead has been gaining attention for its relatively higher boiling point, among other things. On the other hand, a higher melting point makes lead more susceptible to coolant freezing accidents. Surprisingly, LBE has considerably better physical properties than lead, such as a lower melting point (123.5°C). Bismuth does not have any specific advantage in terms of thermophysical properties over other selected metals. According to Table 3.1, tin has the highest boiling point and comparably higher thermal conductivity than any other metal except sodium.

2. Chemical Properties

The phrase "chemical property" is context-dependent but generally refers to a material's behavior at ambient conditions, such as room temperature, atmospheric pressure, or exposure to water. Properties become apparent during a chemical reaction and can be observed by changing a substance's chemical identity. The chemical properties of metals need to be studied when they are considered as reactor coolants. Lead, tin, bismuth, and LBE react differently when they are exposed to oxygen and water.

Sodium, like other Group I[37] elements, is a metal of very high chemical reactivity. When sodium is exposed to air, it burns relatively easily. The result is sodium peroxide, Na_2O_2 and sodium oxide, Na_2O . These chemical reactions are given below:[38]

$$4Na(l)+O_2(g) \rightarrow 2Na_2O+436kj/mole \tag{3.1}$$

$$2Na(s)+O_2(g) \rightarrow Na_2O_2 + 499kj/mole \tag{3.2}$$

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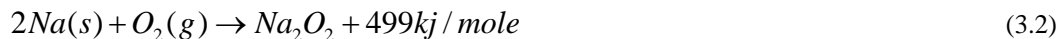
[36] IAEA-TECDOC-1289, p 22.

[37] Group I elements are lithium (Li), sodium (Na), potassium (K), rubidium (Rb), cesium (Cs), and francium (Fr).

[38] Equations (3.1) to (3.4) are taken from, IAEA-TECDOC-1289, p. 22-23.

Sodium reacts rapidly with water. The result is sodium hydroxide, $NaOH$. The reaction is exothermic. The nature of the reaction is highly dependent on the temperature of the reaction and impurities of the sodium.[39] Since the reaction is exothermic and also produces hydrogen gas, the heat produced might ignite an explosion.

$$2Na(l)+2H_2O \rightarrow 2NaOH(aq)+H_2(g)+280kj/mole \qquad (3.3)$$

When sodium reacts with water at high temperatures ($T>450^oC$), the result is expected to be different.[40] The reaction occurs as follows:

$$2Na(l)+2H_2O \rightarrow Na_2O(aq)+H_2(g) \qquad (3.4)$$
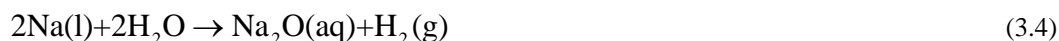
When sodium reacts either with water or air, due to excessive hydrogen gas, the reaction is highly flammable. The hazards of the hydride and oxide produced by the sodium are summarized in Table 3.3. The data in the table is taken from the Material Safety Data Center.[41]

Products	Material Safety Data Sheet
Sodium Oxide (Na_2O)	**Inhalation:** Extremely destructive to tissues of the mucous membranes and upper respiratory tract. Symptoms may include burning sensation, coughing, wheezing, laryngitis, shortness of breath, headache, nausea, and vomiting. **Ingestion:** Corrosive. Swallowing can cause severe burns of the mouth, throat, and stomach. Can cause sore throat, vomiting, and diarrhea.
Sodium Hydride (NaH)	**Inhalation:** Severe irritant. Effects from inhalation of dust or mist vary from mild irritation to serious damage of the upper respiratory tract, depending on severity of exposure. Symptoms may include sneezing, sore throat or runny nose. **Ingestion:** Corrosive! Swallowing may cause severe burns of mouth, throat, and stomach. Severe scarring of tissue and death may result. Symptoms may include bleeding, vomiting, diarrhea, and a fall in blood pressure. Damage may appear days after exposure.

Table 3.3   Hazards of Sodium Reaction Products, Hydride And Oxide

The chemical reactivity of sodium is a serious threat to reactor safety. Any kind of coolant leakage may lead to a major disaster and explosion.

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[39] IAEA-TECDOC-1289, p. 22

[40] Ibid.

[41] http://www.jtbaker.com/msds/englishhtml/s4710.htm, Accessed 06 June 2006, Material Data Safety Sheet, Mallinckrodt Baker Inc. Philipsburg, NJ, USA.

Lead is chemically more stable than sodium. When molten lead interacts with air, it forms lead oxide, PbO. The reaction occurs as follows;

$$2Pb(aq)+O_2(g) \rightarrow 2PbO \qquad (3.5)$$

Under normal conditions, PbO does not react with water. Chemical interaction with water and air without risk of explosion is a prominent passive safety advantage for reactor design.

Qualitatively, the chemical properties of alloys combine most of the properties of their constituent metals. Hence LBE has chemical properties of both lead and bismuth. It is worth noting the chemical reactivity of the LBE coolant to water, steam, and air is relatively low.

Tin is stable in ambient conditions but, if heated to its melting point, it reacts with oxygen to form tin dioxide, SnO_2. The reaction occurs as follows:

$$Sn(l)+O_2 \Leftrightarrow SnO_2 \qquad (3.6)$$

When tin interacts with water, it forms tin dioxide and releases hydrogen. Although the formation of hydrogen may seem disturbing, as in the case of sodium, the formation resulting due to this reaction is far from being either explosive or flammable. Forming out of reaction with water or air, tin dioxide may cause eye and skin irritation, but does not pose serious danger.

Understanding possible chemical reactions of selected reactor coolants with water and air provides invaluable information about the design and safety measures of the nuclear reactors. Findings regarding the chemical reactivity of selected coolants are summarized in Table 3.4.

Coolants	Chemical Reactivity
Sodium (Na)	Highly Reactive
Lead (Pb)	Inert (Caution: Lead Fume Poisoning)
Bismuth(Bi)	Inert
Lead-Bismuth Eutectic(LBE)	Inert
Tin (Sn)	Inert

Table 3.4   Chemical Reactivity of Selected Coolants

B.   MATERIAL COMPATIBILITY AND CORROSION

The coolant selected for the SSTAR application must be compatible with the cladding and should be reasonably compatible with the fuel so that an in-pile breach could be accommodated without adverse fuel coolant interaction.[42] Corrosion of structural material varies substantially among the selected coolants depending on the structural material used. Structural material here is inclusive of all materials normally in contact with the coolant, including core materials, reactor vessel, vessel internal structures, heat exchanger, pumps, valves, etc.[43]

According to E.C. Miller's comprehensive study, there are seven categories of corrosion relevant for liquid metals.[44] Table 3.5 demonstrates the categories in which a mechanism of liquid metal corrosion can be classified.

	Categories of Material Corrosion caused by Liquid Metals		
1	Solution Attack	5	Corrosion Erosion
2	Direct Alloying	6	Thermal Gradient Transfer
3	Inter-angular Penetration	7	Concentration Gradient Transfer
4	Corrosion by Contaminants		

Table 3.5   Categories of Material Corrosion Caused by Liquid Metals

Three of the seven categories need further attention: solution attack, inter-angular penetration, and direct alloying. Solution attack, wherein the liquid metal simply dissolves the metal away at its surface at some rate, is pervasive for the materials addressed here. It is often reported in terms of mass of metal removed per unit surface area of specimen over the test duration or directly in terms of depth of material removed per year of service.[45]

Inter-angular penetration occurs when liquid metals selectively react with minor constituents of the materials they interact with. Selective reaction phenomena are

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[42]Alan E. Walter and Albert B. Reynolds, "Fast Breeder Reactors", p. 441.

[43] Bruce W. Spencer, "Rush to The Heavy Liquid Metal Coolants", 8th International Conference on Nuclear Engineering, April 2-6 2000.

[44] Miller, E. C., 1952, "Corrosion of Materials by Liquid Metals" Chapter 4, Liquid Metals Handbook, R. N. Lyon.

[45] Bruce W. Spencer, "Rush to The Heavy Liquid Metal Coolants", 8th International Conference on Nuclear Engineering, April 2-6 2000.

important in the selection of coolant and structural material. Where sodium is highly corrosive for carbon, lead is safe to interact with such a metal.

Most of the information we have about the compatibility of liquid metals with structural materials dates back to the thorough study funded by the U.S. Navy and conducted by Miller in the 1950s. He uses the mil (0.001 inch) as a primary unit to define the compatibility of liquid metal coolants and materials. Miller explores the material behavior for three different temperatures (300°C, 600°C, and 800°C) and rates them as **POOR** (More than 10 mil per year), **LIMITED** (1 to 10 mil per year), and **GOOD** (less than 1 mil per year). Table 3.6 provides a summary of compatibility information for some common reactor materials and selected liquid metals.

			Na	Pb	Bi	LBE	Sn
FEROUS METALS	Pure Iron	800 C	GOOD	LIMITED	GOOD	UNKNOWN	UNKNOWN
		600 C	GOOD	GOOD	GOOD	GOOD	POOR
		300 C	GOOD	GOOD*	GOOD	GOOD	LIMITED
	Carbon Steel	800 C	POOR	LIMITED	UNKNOWN	UNKNOWN	UNKNOWN
		600 C	LIMITED	GOOD	GOOD	LIMITED	POOR
		300 C	GOOD	GOOD*	GOOD	GOOD	LIMITED
	2–9% Chromium Steel	800 C	LIMITED	LIMITED	LIMITED	GOOD	UNKNOWN
		600 C	GOOD	GOOD	GOOD	LIMITED	UNKNOWN
		300 C	GOOD	GOOD*	GOOD	LIMITED	UNKNOWN
NON FERROUS METALS	Aluminum	800 C	UNKNOWN	UNKNOWN	UNKNOWN	UNKNOWN	UNKNOWN
		600 C	POOR	LIMITED	POOR	UNKNOWN	UNKNOWN
		300 C	POOR	GOOD	GOOD	GOOD	UNKNOWN
	Beryllium	800 C	LIMITED	LIMITED	GOOD	LIMITED	UNKNOWN
		600 C	GOOD	GOOD	GOOD	GOOD	UNKNOWN
		300 C	GOOD	GOOD*	GOOD	GOOD	GOOD
	Nickel and Nickel Alloys	800 C	GOOD	POOR	POOR	POOR	UNKNOWN
		600 C	GOOD	POOR	POOR	POOR	POOR
		300 C	GOOD	POOR	POOR	POOR	POOR
	Copper	800 C	POOR	POOR	POOR	POOR	UNKNOWN
		600 C	GOOD	POOR	POOR	POOR	POOR
		300 C	GOOD	LIMITED*	LIMITED	POOR	POOR
PRYEX GLASS		800 C	UNKNOWN	UNKNOWN	GOOD	GOOD	UNKNOWN
		600 C	POOR	POOR	GOOD	UNKNOWN	POOR
		300 C	LIMITED	GOOD*	GOOD	UNKNOWN	GOOD

Table 3.6   Resistance of Some Materials to Liquid Metals at Various Temperatures[46]

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[46] Miller, E. C., 1952, "Corrosion of Materials by Liquid Metals" Chapter 4, Liquid Metals Handbook.

As we see in Table 3.6, the resistance of materials to liquid metals changes significantly with temperature. Sodium has favorable corrosion behavior compared to other selected liquid metals. By now, comprehensive studies have been performed and reliable industrial experience has been gained on material corrosion with sodium.[47] Meanwhile, material resistance to lead, bismuth, and LBE varies substantially with different temperatures and materials.

Unlike other coolants, tin poses another challenge to the problem of selecting coolants for power reactors. We hardly understand material behavior when interacting with tin. We lack the half-century operating experience that we have with LBE and sodium reactors. For this reason, some material combinations with tin in Table 3.7 are rated as *unknown*. The compatibility and corrosion behavior of selected coolants are individually discussed below:

1. Sodium (Na):

Based on reactions with structural materials, sodium emerges as the most compatible candidate coolant. Even at high temperatures, sodium provides favorable resistance rates with many metals. Pure iron, chromium steels, stainless steels, austenitic stainless steels, zirconium, nickel and its alloys, and graphite all promise good resistance between the melting and boiling temperatures. Austenitic stainless steels (type 304 and 316) have been used in the U.S. LMFBR program owing to its good compatibility with the coolant and its good performance under irradiation.[48] Recently, the ferritic stainless steel HT-9 has also received attention owing to its superior performance.[49]

With sodium, as with other liquid metals, the corrosion rate depends not only temperature but also on many other factors, such as coolant velocity, the content of impurities such as oxygen, and the solubility of metals. Type-316 stainless steel (316 SST) has been excessively studied due to its good compatibility. Figure 3.2 demonstrates corrosion data of 316 SST with different oxygen impurities and temperatures.
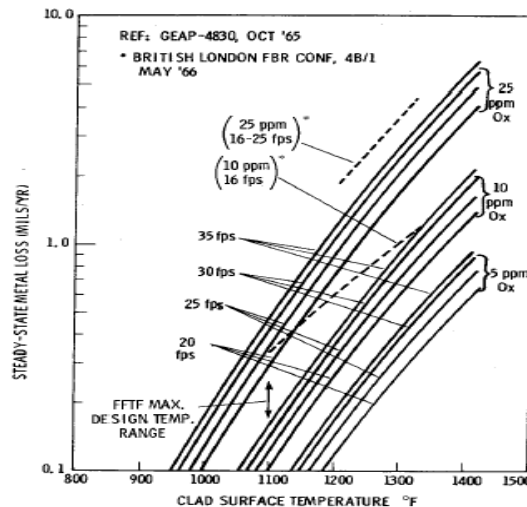
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[47] IAEA-TECDOC-1289, p.32.

[48] Bruce W. Spencer, "Rush to the Heavy Liquid Metal Coolants", 8th International Conference on Nuclear Engineering, April 2-6 2000.

[49] Leggett, R. D., and Walters, L. C., 1993, "Status of LMR Fuel Development in the United States", Journal of Nuclear .Materials, 204, 23-32.

Figure 3.2   Corrosion data of 316 SST (Stevens, 1970)

Figure 3.2 is important because it shows that the corrosion rate increases with higher temperatures and that a higher level of oxygen impurity also leads to higher corrosion rates. However, stainless steels also permit higher oxygen content with sodium than is compatible with lower alloy steels.[50]

In general, sodium is very compatible with many structural materials. Operational experience with a number of LMFBRs during a 20–30-year period has shown that sodium is practically non-corrosive with respect to stainless steel when the content of impurities, mainly oxygen and carbon, are held at an acceptably low level by cold traps.[51]

2.   Lead (Pb)

Lead is highly corrosive to many of the possible structural materials. Lead exhibits a strong erosion/ corrosion effect on structural materials, such as material dissolving, embrittlement, thermal transport of mass, and inter-angular penetration of lead.[52]

The resistance of materials to corrosion by molten lead varies in great degree in relation to temperature and solubility of the material. The main contributor to material corrosion is the dissolution of contact materials and their components in these coolants.

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[50] Bernard Kopelman, "Materials for Nuclear Reactors", 1959, p.331.

[51] IAEA-TECDOC-1289, p. 32.

[52] Ibid.

The solubilities of some metals in molten lead, bismuth, and LBE are given in Table 3.7.[53]

Material	Pb		Bi		LBE	
	ppm	Temp °C	ppm	Temp °C	ppm	Temp °C
Cu	3600	500	7200	500	14800	500
Fe	6.7	700	33	700	125	700
	2.6	600	10	600	50	600
Cr	730	600	--	--	150	600
Ni	5370	600	--	--	66000	600
Co	10.2	600	--	--	450	600

Table 3.7 Solubilities of Cu, Fe, Cr, Ni, and Co in Pb, Bi, and LBE

According to Table 3.7, the solubility of copper and nickel in molten lead is substantially higher than that of iron, chromium, and cobalt. Although the data indicates that iron offers low solubility in lead, the level is still too high to permit lead to contact steel without some form of protection. The relatively high solubility of nickel in lead also necessitates the use of inhibitors. In this case, the oxygen content of the molten lead plays an important role. It forms oxide layers on the surface of the contact material and reduces the corrosion rate since it prevents direct contact. The corrosion resistance of metals in molten lead essentially depends on the concentration of dissolved oxygen.[54]

The oxygen level must be carefully maintained by a technique called active oxygen control.[55] If not enough oxygen is present in the coolant, the protective oxide films on the steel surfaces do not form, thereby permitting dissolution corrosion. If too much oxygen is present, exceeding solubility, then solid lead and/or bismuth oxides can form from the overabundance of oxygen.[56] Such oxides contaminate the coolant and can precipitate out in cool regions, possibly clogging the flow of coolant.[57] The main idea behind active oxygen control is that by properly controlling oxygen levels, the reduction process can be kept at equilibrium between the formation and dissolution of oxides.

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[53] J.R.Weeks, "Lead, Bismuth, Tin and Their Alloys As Nuclear Coolant", Nuclear Engineering. and Design, 1971.
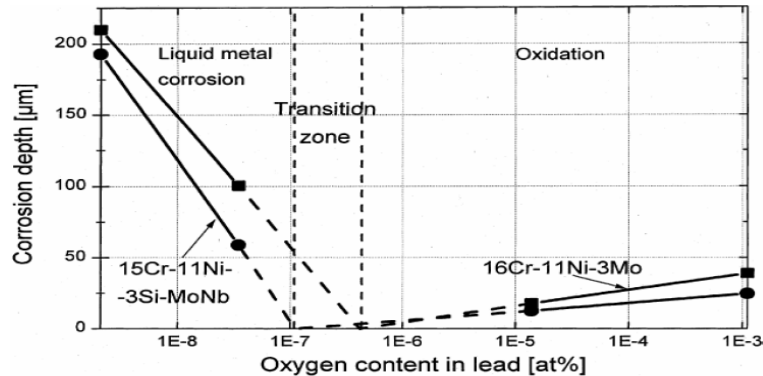
[54] IAEA-TECDOC-1289, p.32.

[55] Daniel Koury, "Investigation of the Corrosion of Steel by Lead-Bismuth Eutectic (LBE) Using Scanning Electron Microscopy and X-Ray photoelectron Spectroscopy", 1998.

[56] Ibid.

[57].Ibid.

The effect of the formation of a protective oxygen layer on the corrosion behavior of steels in molten lead is shown in Figure 3.3.

Figure 3.3 Effect of Formation of Protective Oxygen Layer on Corrosion Behavior of Steels in Molten Lead[58]

When the oxygen concentration is above 10^{-6}, a protective oxide layer is formed between molten lead and steels. Since this oxide layer prevents direct contact between coolant and material, corrosion depth significantly decreases at a thickness above $10^{-6.}$

According to Table 3.7, ferrous steels, such as pure iron, carbon steel, and chromium steel, have good resistance to molten lead attack below $600^{o}C$. Non-ferrous metals show varying resistance to lead attack due to their different solubility behaviors in molten lead. Tantalum and columbium have good resistance to lead attack at $1000^{o}C$.[59] Where beryllium has good resistance to lead attack up to $600^{o}C$, nickel and its alloys have poor resistance to attack by lead at the melting point of lead.[60]

The Russians have designed BREST 300 and BREST 1200, 300 Mwe, and 1200 MWe reactors, which are lead-cooled.[61] They have acknowledged the corrosion and compatibility issues of molten lead with structural material. The necessity of a protective

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[58] Gorynin, I.V., Karzov, G.P., Markov, V.G., Lavrukhin, V.S., Yakovlev, V.A., "Structure Materials for Power Plants with Heavy Liquid Metals as Coolants," Proceedings: Heavy, Liquid Metal Coolants in Nuclear Technology, Vol. 1 (Hlmc-98), Obninsk, Russia.

[59]"Liquid Metals Handbook", Atomic Energy Commission, Department of Navy, June 1952.

[60] Ibid.

[61] http://www.nikiet.ru/eng/structure/mr-innovative/brest.html Accessed 24 November 2006, Department of Development Institute of Power Engineering, Russian Federation.

layer between the structural material and the molten lead emerges as a disadvantage compared to sodium, which requires no corrosion protection measures other than impurity control.

3.   Lead Bismuth Eutectic (LBE):

As with molten lead and bismuth, the corrosion of structural materials in LBE occurs mainly through dissolution of materials when they contact liquid LBE.   The solubilities of chromium, nickel, and iron are given in Table 3.8.

Nickel has greater solubility in LBE than any other metal listed in the table. However, solubility data is given only for a single temperature figure. Solubility data for nickel, iron, and chromium are given for different temperatures in Figure 3.4.
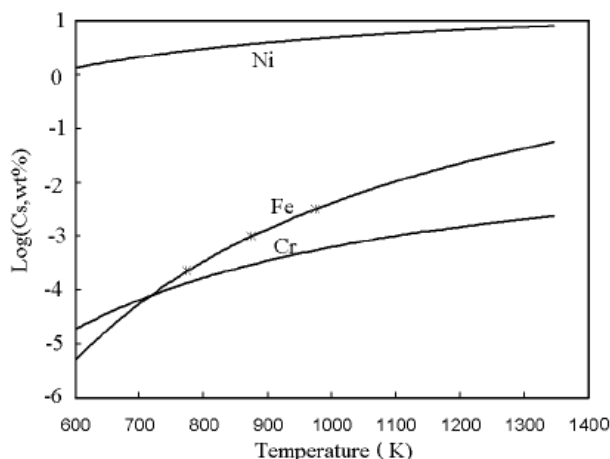
Figure 3.4   Solubility of Fe, Cr, and Ni in LBE at Varying Temperature (Weeks, J., Romano, *Corrosion* 25, 1969)

The solubility of iron and chromium dramatically increase with rising temperature. Moreover, Figure 3.4 puts the high solubility of nickel in LBE in perspective.[62] Thus, the nickel content of any structural material in contact with LBE might lead to serious corrosion problems.

The solubility of a metal in liquid lead-bismuth alloys depends on the compositions of the liquid alloy.[63] Figure 3.5 demonstrates the solubility behavior of iron in LBE, with varying lead and bismuth compositions, at different temperatures. As seen,

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[62] Jinsuo Zhang, Ning Li, "Review of Studies on Fundamental Issues in LBE Corrosion**",** LA-UR-04-0869.Department of Energy under contract number W-7405-ENG-36.

[63] Ibid.

the solubility of iron increases with temperature and with higher bismuth compositions. LBE (44.5 wt% Pb and 54.5 wt% Bi) appears to be less corrosive than pure bismuth but more corrosive than pure lead.
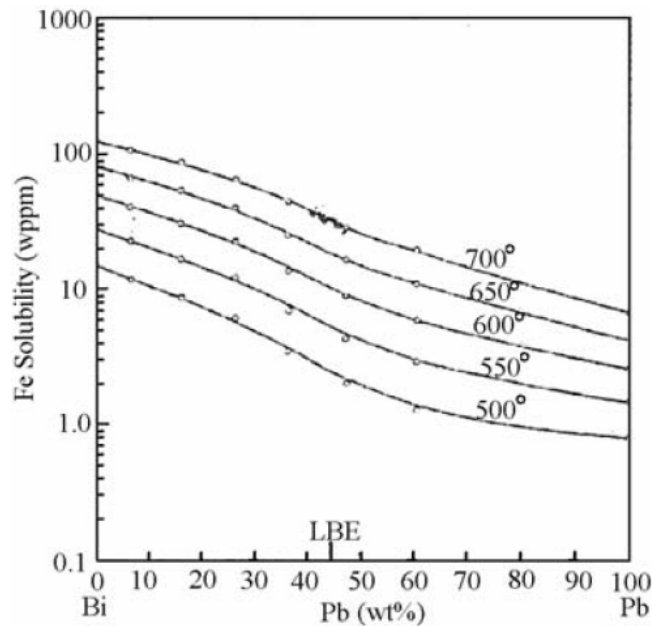
Figure 3.5   Solubility Of Fe in Lead-Bismuth Composition at Different Temperatures. (Weeks, Romano, *Corrosion* 25,1969)

The high solubility of structural materials in LBE requires the use of a protective layer between LBE and contact material. Oxygen, with molten lead, and zirconium with liquid bismuth, both promise viable solutions to the dissolution corrosion phenomenon. Either oxygen or zirconium can be used in LBE for the same purpose.

After protective films are formed, they should be monitored and restored in case of deformation that exposes material to direct contact with LBE. Thus, inhibitors must remain in the liquid at all times to ensure the restoration of the films in spalled areas.

Iron, carbon, and high-chromium steel have good resistance to liquid LBE at temperatures up to 600°C. Stainless steels have different corrosion resistance, which is highly temperature-dependent. For instance, type 302 and similar 18 Cr-8 Ni austenitic stainless steels have good resistance to attack by LBE at 230°C, limited resistance at 570°C, and poor resistance at 1000°C. Beryllium, a non-ferrous metal, has good resistance to LBE attack up to 700°C. Nickel, copper, and their alloys have so poor resistance to LBE attack that they cannot be used without some form of protection.

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4.   Tin (Sn)

Liquid tin is one of the most aggressive liquid metals; it reacts with steels to form $FeSn_2$ (well-known tinning reaction) so that molten tin or liquid alloys containing tin, such as Bi-Pb-Sn ternary eutectic, cannot be used as coolants in ferritic and austenitic systems at any temperatures substantially above their melting points.[64]   This fact seriously limits the available structural material that is compatible with liquid tin. Only some refractory materials, such as titanium, tungsten, molybdenum, tantalum, and columbium, have sufficient corrosion resistance to attack by liquid tin.[65]

The other disadvantage of liquid tin is the lack of relatively new data on corrosion. All information in the literature about liquid lead dates back to the 1950s. Arguably, violent corrosion behavior with contact to many potential structural materials is a primary drawback of tin as a coolant in general.

C.   COST OF LIQUID METAL COOLANTS

The financial cost of liquid metals is also a factor to consider since a great amount of metal will be used and wasted during operations.[66] For example, bismuth is a scarce material; the capital cost of the LBE alloy increases with the percentage of bismuth.

In order to estimate the capital cost of selected coolants, the following assumptions are made:

- ➢ Market cost of lead[67](C): $1/kg
- ➢ Market cost of bismuth[68](C): $7/kg
- ➢ Market cost of LBE (54.5 wt%Bi and 44.5 wt%Pb)(C): $4.5/kg
- ➢ Market cost of tin[69](C): $0.64/kg
- ➢ Market cost of sodium[70](C): $0.3/kg

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[64]J.R.Weeks, "Lead, Bismuth, Tin and Their Alloys as Nuclear Coolant", Nuclear Engineering and Design, 1971.

[65] Ibid.

[66] Buongiorno, Jacopo, "Conceptual Design Of A Lead-Bismuth Cooled Fast Reactor With In-Vessel Direct-Contact Steam Generation" Thesis (Ph.D.)--MIT, Dept. of Nuclear Engineering, 2001.

[67] http://mineral.usgs.gov Accessed 22 October 2006, United States Geological Survey.

[68] Ibid.

[69] Ibid.

[70] http://en.wikipedia.org/wiki/Sodium Accessed 22 October 2006, Wikipedia online Dictionary.

➢ Capacity factor(f)[71]:0.85

➢ Mass per unit electric power(m):5 kg/kWe

➢ Annual capital carrying charge factor(κ):10%

The capital cost of coolant, c_e (mills/kWh), can be calculated as;

$$c_e = \frac{m_{coolant} * C_{coolant}}{8760\,f} \kappa \qquad\qquad (3.5)$$

Calculated capital costs are given in Table 3.8.

	Na	Lead	LBE	Tin
c_e (mills/kWh)	0.021	0.072	0.326	0.046

Table 3.8   Capital Costs of Selected Coolants

In addition to Table 3.8, Figure 3.6 is also helpful in appreciating the varying costs of different coolants and also calculates the capital costs of different coolants.
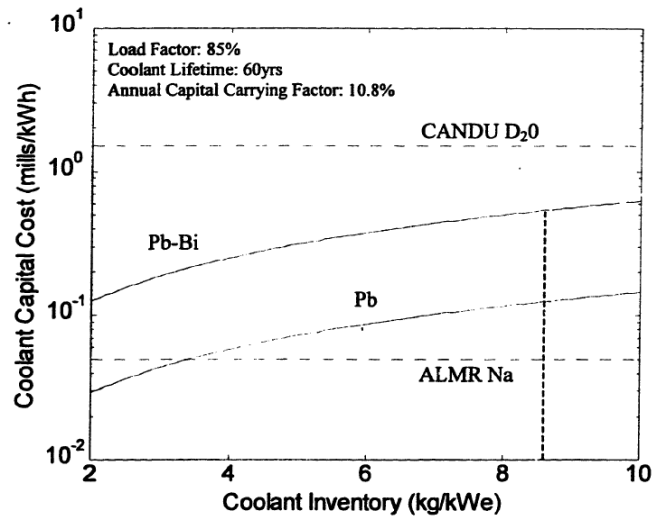
Figure 3.6   The Capital Cost of Heavy Liquid-Metal Coolants

In Figure 3.6[72], the capital costs of heavy water ($250/Kg by Hejzlar, 1994) and sodium, lead, and LBE are shown as a function of mass per unit of electric power. It can be seen that sodium is the least costly coolant. Although the cost of lead is comparable to

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[71] Calculation is based on the assumption of reactor is operated %85 of its full capacity.

[72] Buongiorno, Jacopo, "Conceptual Design of a Lead-Bismuth Cooled Fast Reactor with In-Vessel Direct-Contact Steam Generation" Thesis (Ph.D.)--MIT, Dept. of Nuclear Engineering, 2001.

that of sodium, the cost of LBE is far higher. Due to the intensive work needed for isotropic separation, CANDU, a heavy cooled reactor, is the least efficient.

IV. COOLANT ACTIVATION

A. PROBLEM STATEMENT

A major concern related to the implementation of any neutron-producing device is the induced radioactivity. Particularly for my scope of study, coolant activation in nuclear reactors is an important research area due to the need to ensure safe operations and effective waste management.

Radioactivity in coolants originates in several ways: by neutron activation of coolant, by activation of impurities contained in the coolant, and by picking up particles of activated structural material in the reactor.[73] The activity acquired by a coolant by these processes depends on the flow pattern, its residence time in and out of a neutron flux, and particle cross sections.

During my research, with the help of Dr. Craig F. Smith and scientists at the Lawrence Livermore National Laboratory, we contacted different research institutes all around the world, including IPPE in Russia, the Nuclear Energy Agency (NEA) in France, and the Argonne National Laboratory in the U.S. Surprisingly, we found that there are significant differences in the calculation of coolant activation and decay heat load, particularly for LBE coolant.

Through personal contact via Dr. Neil Brown at the Lawrence Livermore National Laboratory, we learned that Russian scientists with considerable experience with LBE coolant technology calculated that the Po-210 heat decay in LBE cooled reactor would be equal to 28% of the heat load 100 hours after shutdown. In addition, studies in the United States and Europe indicated divergent projections of the Po-210 generation in LBE systems.

Given the fact that there are substantial differences among the different calculations, it seemed worthwhile to go further and calculate the activation of radioactive nuclides not only for LBE coolant but also for lead, sodium, and tin.

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[73] Lamarsh, John, "Nuclear Engineering", 2001, p. 599.

The analytical tool used in this assessment is ORIGEN. ORIGEN[74] uses the automatic rapid processing (ARP) interpolation methodology for generating problem-dependent cross section libraries. It computes time-dependent concentrations and radiation source terms of a large number of isotopes, which are simultaneously generated or depleted through neutronic transmutation, fission, and radioactive decay.

B. ORIGENARP MATRIX SOLUTION TO THE BATEMAN EQUATION

In determining the time dependence of nuclide concentrations, OrigenArp[75] is primarily concerned with developing solutions for the following equation:

$$\frac{d\mathrm{N}_i}{dt} = \text{Formation Rate - Destruction Rate - Decay Rate} \tag{4.1}$$

Specifically, the time rate of change of the concentration for a particular nuclide, N_i, and numerical solution to the Bateman equation can be written:

$$\frac{d\mathrm{N}_i}{dt} = \sum_j \gamma_{ij}\sigma_{f,j}N_j\phi + \sigma_{c,i-1}\mathrm{N}_{i-1}\phi + \lambda_i'\mathrm{N}_i' - \sigma_{f,i}N_i\phi - \sigma_{c,i}N_i\phi - \lambda_i\mathrm{N}_i \tag{4.2}$$

Where (i=1,…,I)

$\sum_j \gamma_{ij}\sigma_{f,j}N_j\phi$ is the yield rate of N_i due to fission of all N_j;

$\sigma_{c,i-1}\mathrm{N}_{i-1}$ is the transmutation into N_i due to radioactive neutron capture by nuclide N_{i-1};

$\lambda_i'\mathrm{N}_i'$ is the rate of formation of N_i due to radioactive decay of nuclides N_i';

$\sigma_{f,i}N_i$ is the destruction rate of N_i due to fission;

$\sigma_{c,i}N_i$ is the destruction rate of N_i due to all forms of absorption other than fission; and

$\lambda_i\mathrm{N}_i$ is the radioactive decay rate of N_i

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[74] The version of ORIGEN used in this work is ORIGEN-Arp, or ORIGEN with Autonomous Rapid Processing.

[75] Equations and information are taken from "Origen-Arp: Automatic Rapid Processing For Spent Fuel Depletion, Decay, And Source Term Analysis" by I. C. Gauld, S. M. Bowman, J. E. Horwedel, L. C. Leal and "Origen-S: Scale System Module to Calculate Fuel Depletion, Actinide Transmutation, Fission Product Build up and Decay, and Associated Radiation Source Terms" by I. C. Gauld, O.W. Hermann and R.M. Westwall.

OrigenArp uses the matrix exponential method in solving complex numerical equations, such as the Bateman equation derived above. For all nuclides, N_i, equation (4.2) represents a coupled set of linear, homogenous, first-order differential equations with constant coefficients. Therefore, equation (4.2) can be written in matrix notation as:

$$\dot{N} = A\,N \tag{4.3}$$

In this formulation N is a vector of nuclide concentrations and A is the transition matrix containing the rate of coefficients for radioactive decay and neutron absorption. The solution to the system of equations equation is going to be:

$$N = \exp(A\,t)\,N(0) \tag{4.4}$$

Where $N(0)$ is a vector of initial nuclide concentrations.

If we expand the exponential expression, $\exp(At)$, as below:

$$\exp(At) \equiv I + At + \frac{(At)^2}{2!} + \frac{(At)^3}{3!} + \ldots = \sum_{m=0}^{\infty} \frac{(At)^m}{m!} \tag{4.5}$$

and substitute the matrix exponential function in Equation (4.5), we have:

$$N = \left[I + At + \frac{(At)^2}{2!} + \frac{(At)^3}{3!} + \ldots \right] N(0) \tag{4.6}$$

This result is a series of terms that arise from the successive post-multiplication of the transition matrix by the vector of nuclide concentration produced from the computation of the previous term.

So far in the discussion of the analytical model applied in the OrigenArp, it has been assumed that the equation is homogeneous. This equation is applicable to fuel burn and decay calculations; therefore, its solution is of primary interest for our calculations. Although OrigenArp is also capable of solving heterogeneous equations with externally imposed nuclide concentrations, such application is beyond the scope of this study.

C.   WORK DESCRIPTION

Calculations of activation were performed using the OrigenArp computer code. Although analytical methods significantly reduced limitations and increased accuracy and

speed in comparison with manual calculations, we still, to a certain extent, need to simplify inputs by making some assumptions, such as those related to flux distribution and material concentration.

When calculating the activation of coolant, the neutron flux is as important as the coolant material. However, neutron flux varies from location to location within the reactor. In actuality, even for the same material, production rates may differ from point to point inside core. A proposed design schematic of a 30-Mwe SSTAR core is shown in Figure 4.1.
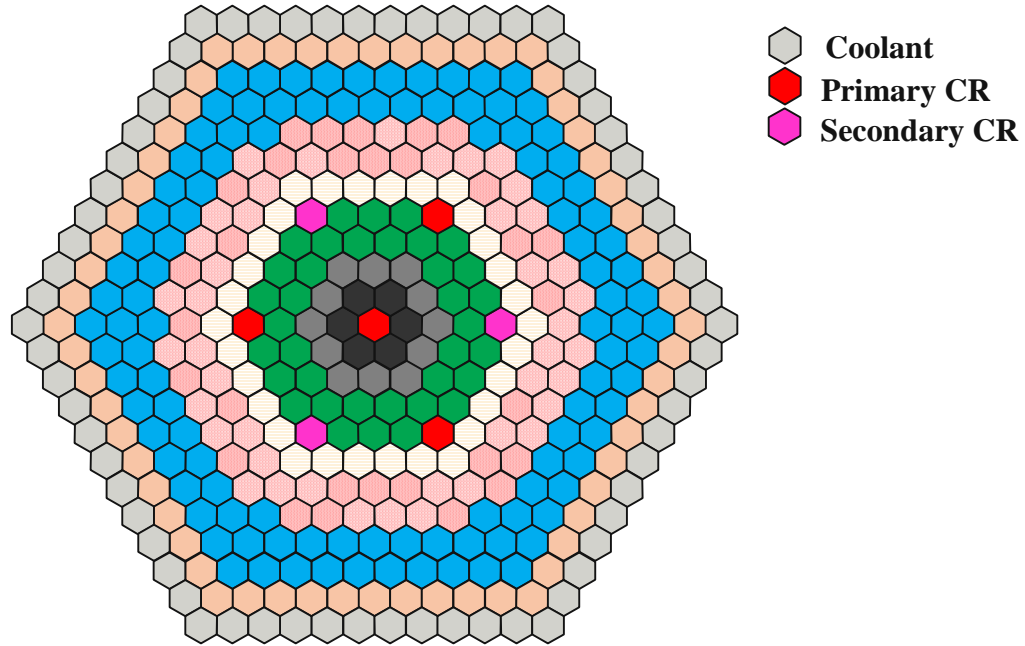
Figure 4.1   SSTAR Core Design[76]

Coolant activation calculations are carried out under the following conditions:

- The neutron spectrum characteristics of the central area of the SSTAR are used in the calculations. The spectrum is in the 20ev to 15Mev neutron energy range, with wide maximum in the 0.1 to 2.0 Mev region. Integral neutron flux with energy $E_{n\,min}$ and $E_{n\,max}$ is: $\phi = \int_{E_{n\,min}}^{E_{n\,max}} \phi(E)dE = 5.40*10^{14}\,n/cm^2/s$ [77].

<hr>

[76] Personal contact with Dr.Won Sik Yang of Argonne National Laboratory on August 3, 2006.

[77] Ibid.

- The annual neutron fluence is $F = 1.70 * 10^{22} \, n/cm^2$, while maximum fluence, corresponding to an exposure time of 1825 days (5 years), is $8.514 * 10^{22} \, n/cm^2$.

- The initial mass of the coolant material considered for the calculation is equal to 1000 grams.

- The coolant is activated only while it flows inside the core.

- The coolant volume fraction of the active core is 0.278.

- The OrigenArp computer code is used to analyze the activity of coolants and nuclear reaction products from irradiation of different target coolants in the SSTAR neutron spectrum.

- The libraries generated for OrigenArp are based primarily on ENDF/B-VI, FEDL, and EAF evaluations[78]. These provide data for about 800 isotopes. Assembly-specific time-dependent cross sections, developed from lattice physics calculations, are derived from multi-group ENDF/B-VI cross sections for about 220 isotopes.

D. MODIFICATIONS IN ORIGENARP INPUT

The OrigenArp system does not contain all the options in the more complete version of ORIGEN known as ORIGEN-S. This is a design limitation that was chosen to simplify running ORIGEN-Arp for the average user. The currently available version of OrigenARP provides menus and toolbars that assist the user in preparing input files for reactor options, including a number of pressured water reactors (PWRs), a boiling water reactor (BWR), and a CANDU reactor.[79]

However, for fast reactor calculations, one has to make some changes to the text editor of the input file. Below are the modifications made for completing the activation calculations.

- Arp section of the input file deleted up to #ORIGEN

- Set array 0$$ entry 4, NDSET, to 1

- Set array 3$$ entry 1, to 28[80]

---

[78] ENDF-VI, FEDL, EAF are different cross section libraries which include multi-group cross sections of more than 200 isotopes.

[79] "OrigenArp User Manual", Scale 5, Oak Ridge National Laboratory, 2004

[80] Personal contact with Scale help center, Dr. Ian Gauld, 27 September 2006

- Set array 4$$ entry 1, 2 and 3 to 1.0 {THERM (Thermal), RES (Resonance) and FAST (Fast) have meaning for only thermal reactors. In this case THERM, RES, and FAST are set to 1.0}

- Array 56$$ (Sub case Control Constants entry 3 needs to be changed to 1. (enables Neutron Flux input)[81]

- Add array 59$$ specifying neutron flux for each irradiation cycle.

- Array 73$$ needs to be changed (remove fuel material)

- Array 74$$ needs to be changed (remove fuel concentrations)

- Array 73$$ needs to be changed (remove fuel actinide library entries)

- Set LIBUNIT to 21 for preventing PLOTOBUS error.(Since we use ORIGEN-s interface, we need to change it in order to see plot)[82]

E.   RESULTS RELATED TO APPLICATION OF ORIGEN-ARP

1.   Lead (Pb)

Natural lead is the mixture of four stable isotopes: Pb-208, Pb-207, Pb-206, and Pb-204. Pb-208 accounts for more than half of this mixture. The composition of natural lead is given as follows: 52.35% (Pb-208), 22.08% (Pb-207), 24.14% (Pb-206), and 1.42% (Pb-204). For lead coolant, the natural abundance of isotopes with corresponding percentages are used in the simulation.

When LBE or lead coolant is used as a coolant medium, attention is paid to the generation of alpha active polonium formed in the coolant under irradiation. Handling radioactive coolant requires special measures for radiation safety. Although in normal operations, in which the coolant loop is sealed, polonium is not hazardous. The danger arises under the primary loop break, for repair work or nuclear fuel reloading.[83]

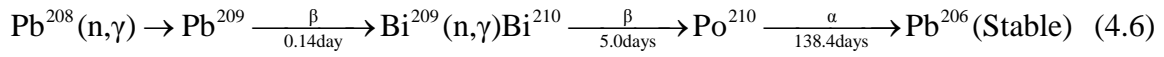[81] Personal contact with Scale help center, Dr. Ian Gauld, 27 September 2006.

[82] Ibid.

[83] Buongiorno, Jacopo, "Conceptual Design of a Lead-Bismuth Cooled Fast Reactor with In-Vessel Direct-Contact Steam Generation" Thesis (Ph.D.)--MIT, Dept. of Nuclear Engineering, 2001.

Polonium has no stable isotopes. However, Po-210, Po-214 and Po218 do naturally occur in the day chain of U-238(GE, 1996). The isotope of interest in our case, Po-210, emits alpha particles of 5.3Mev. In a lead-cooled reactor, the problem of polonium contamination exists because of Bi-209 formation by neutron capture in Pb-208.

Whereas in the LBE reactor, the polonium activity value is determined by the direct activation of Bi-209 isotope, polonium activity in lead is determined by the quantity of Pb-208.

Po-210 is generated as a result of nuclear reaction as below:

$$Pb^{208}(n,\gamma) \rightarrow Pb^{209} \xrightarrow[0.14day]{\beta} Bi^{209}(n,\gamma)Bi^{210} \xrightarrow[5.0days]{\beta} Po^{210} \xrightarrow[138.4days]{\alpha} Pb^{206}(Stable) \quad (4.6)$$
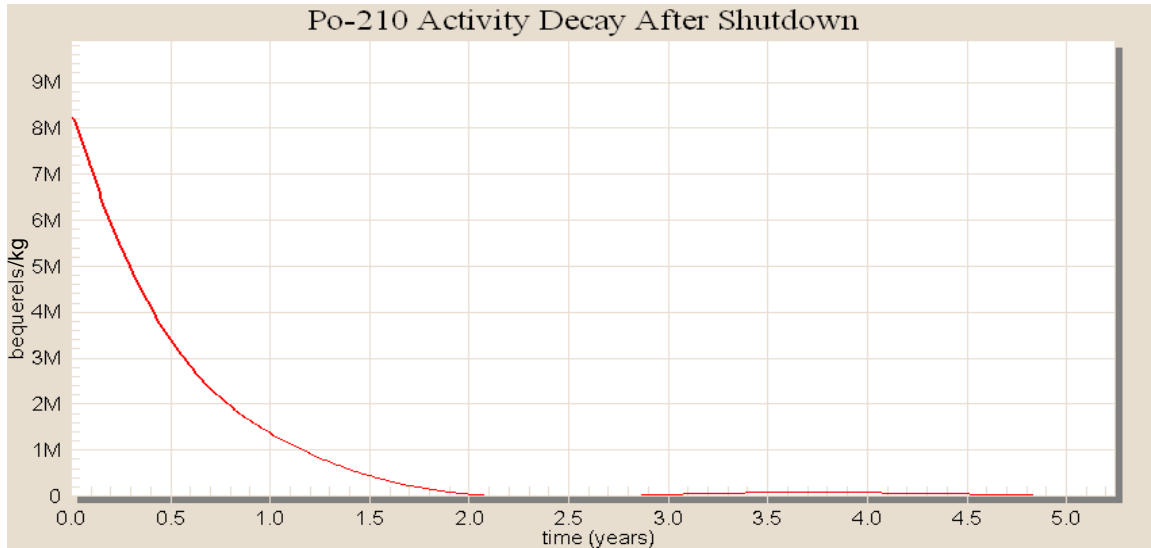
Polonium activation in lead coolant over 1825 days (5 years) of irradiation is given in the plots below:
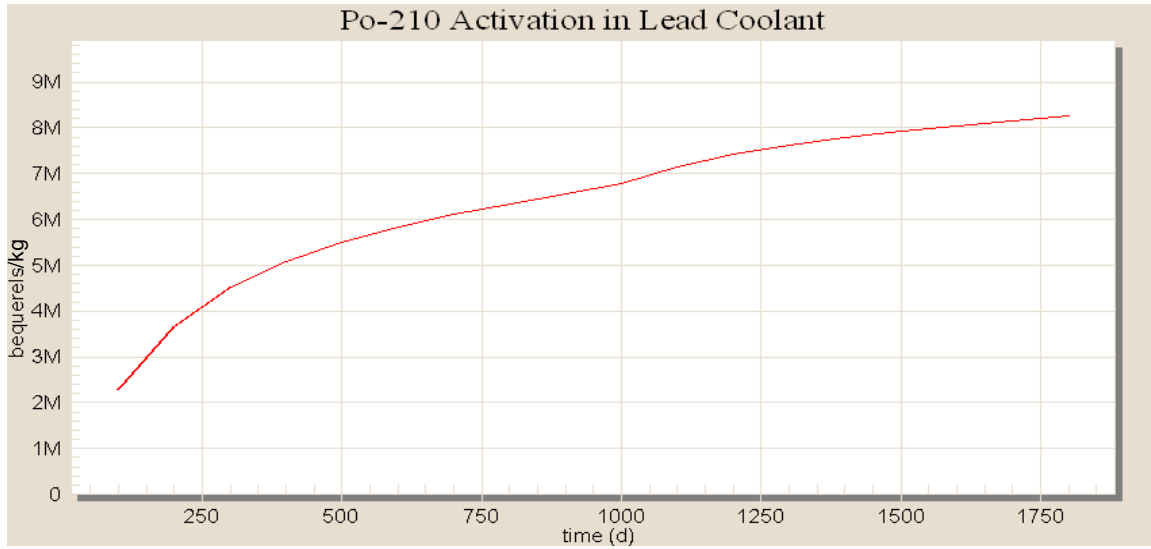
Figure 4.2 Po-210 Activity After 1825 day (5 years) Irradiation in Lead Coolant

Over five year irradiation time, the Po-210 activity in lead coolant reaches $8x10^6$ Bq per kilogram of lead in the reactor. Our simulation results have confirmed that polonium buildup is a real phenomenon, which needs to be dealt within the design phase.

However, Po-210 is a relatively short-lived nuclide. It decays with a half-life of 138.4 days. Figure 4.3 and Figure 4.4, in linear and logarithmic scale, respectively, demonstrate Po-210 decay after shutdown.

Figure 4.3   Po-210 Decay in Lead Coolant (Linear)
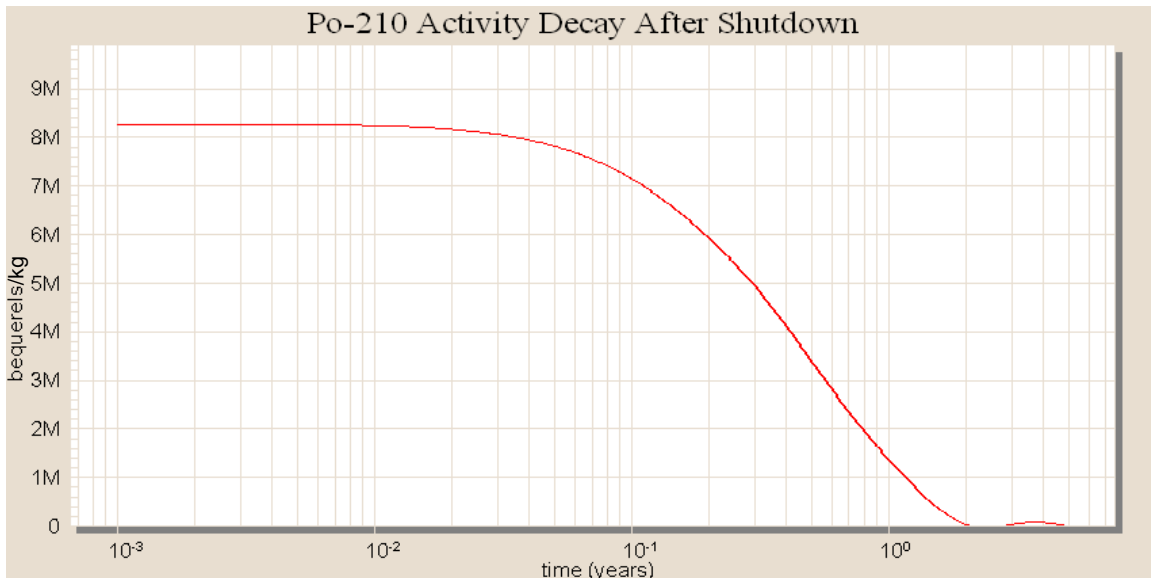
Figure 4.4   Po-210 Decay in Lead Coolant (Logarithmic)

We can see a rapid decay of polonium activity after its half-life in both figures above. Another alpha active radionuclide is Pb-210, with a half-life of 22.26 years ($T_{1/2} = 22.26\,year$). It builds up to 300 Bq per kilogram lead as seen in Figure 4.5. Therefore, it does not pose any safety concern. Pb-210 activity after five years of irradiation is given in Figure 4.5.
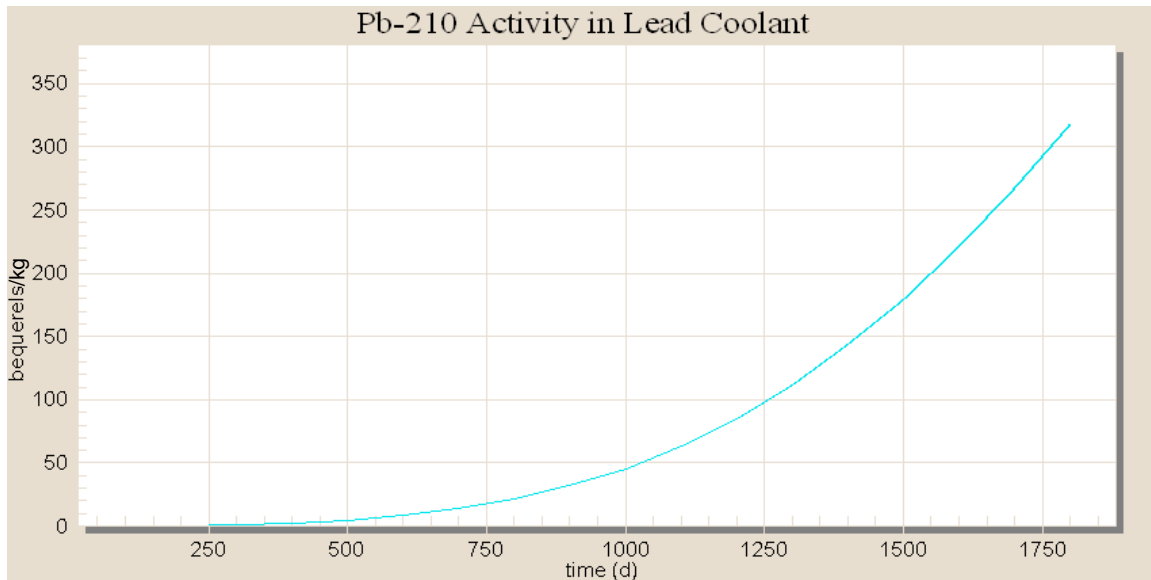
Figure 4.5   Pb-210 Activity after 1825 day (5 years) Irradiation in Lead Coolant

If removing bismuth is not envisaged, a concentration of stable bismuth will increase year by year as a result of nuclear reactions.[84] Hence, Bi-208, a stable isotope, will be available in the coolant even over a longer cooling down period. Figure 4.6 shows the stable activity level of Bi-208 over a five-year cooling down period.
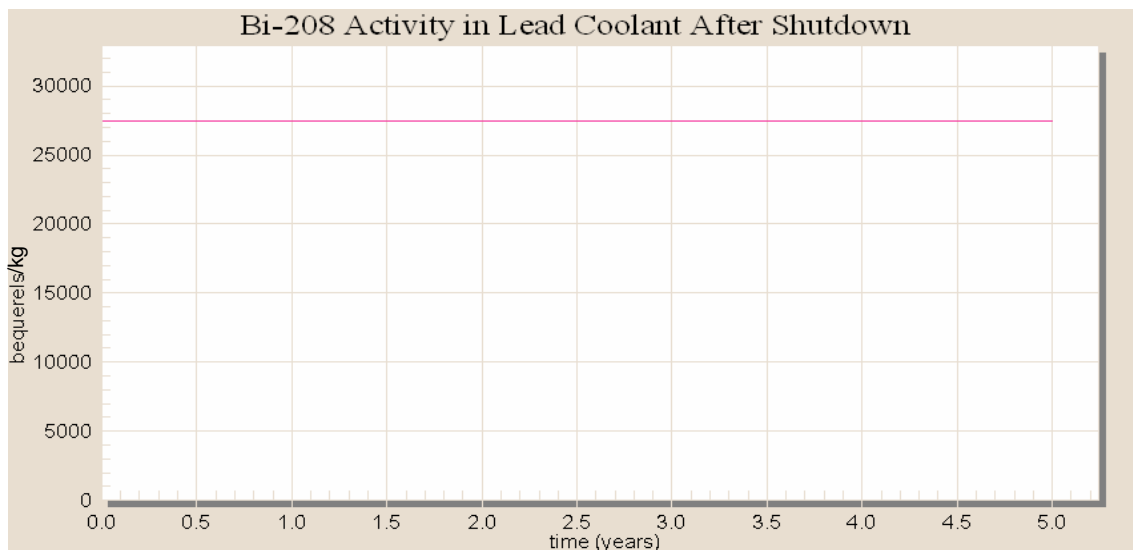

Figure 4.6   Bi-208 Activity in Lead Coolant after Shutdown

[84] G.L. Khorasanov, A.P. Ivanov, A.I. Blokhin, A.L. Shimkevich, M.V.Mikhailyukova. "Lead and Tin Targets for Reducing Polonium Waste", 2002, available from http://www.fjfi.cvut.cz/con_adtt99/papers/Tu-o-e8.pdf, Accessed 01 December 2006, Institute for Physics and Power Engineering, Russia.

Meanwhile, the most important contribution to long-term activity of lead coolant comes from Pb-205 ($T_{1/2} = 1.51*10^7\,y$). Pb-205 is generated from Pb-204(n,γ) and Pb-206(n,2n). Specific β activity of Pb-205 dominates the overall activity of lead coolant in the long term. Although Pb-205 activity does not exceed 300K Bq/Kg, unlike Po-210 and Pb-210, it will not decay in a short time. The decay of Pb-205 activity over 100 years is given in Figure 4.7.
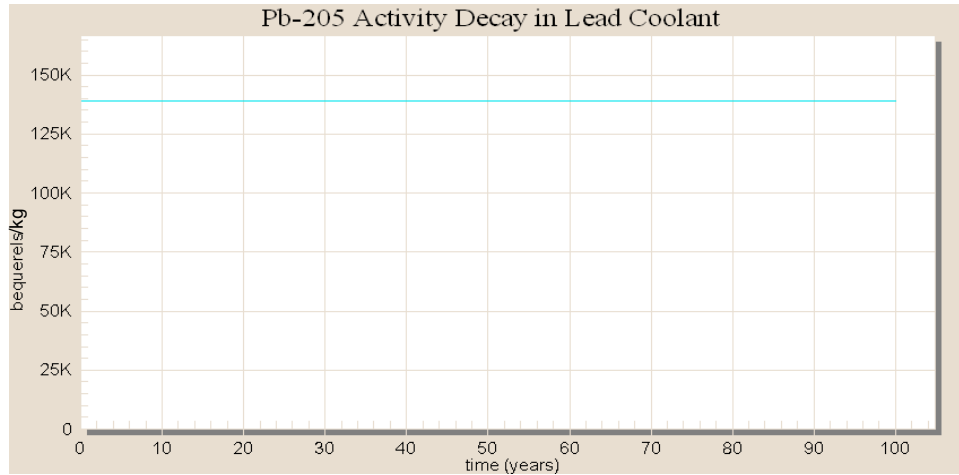
Figure 4.7   Pb-205 Activity in Lead Coolant after Shutdown (100 years)

Important points related to lead coolant activation in the simulation are given below:

- Even if bismuth isotopes are initially absent in the simulation, bismuth is produced within the nuclear reaction of lead isotopes.

- We observed a substantial amount of Po-210 buildup ($8x10^6$ Bq/Kg).

- While Po-210's activity dominates overall activity during a cooling down period of less than one year, due to their long half-lives, Pb-205 and Bi-208 activities will dominate in longer cooling down periods.

2.   **Lead Bismuth Eutectic (LBE)**

LBE has a composition of 44.5 wt% lead and 55.5 wt% bismuth. Bi-209, an isotope of bismuth, is 100% available in nature. In order to simulate the material composition of LBE coolant, I used 555 grams of Bi-209 and 445 grams of lead, with corresponding percentages of their availability in nature.

In an LBE-cooled reactor, Po-210 is generated as a result of following nuclear reaction:

$$Bi^{209} + n \rightarrow Bi^{210} \xrightarrow[5\,days]{\beta} Po^{210} \xrightarrow[138.4\,days]{\alpha} Pb^{206} \tag{4.7}$$
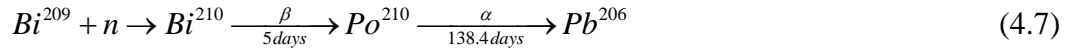
Like the lead coolant, Po-210 activity is the primary concern in LBE coolant. However, while in the case of lead coolant, a main source of Po-210, bismuth, is produced as a byproduct of nuclear reaction, bismuth is initially available in the LBE coolant.

In Chapter II, I considered the neutron capture cross sections of Bi-209 and Pb-208. In fact, the neutron capture cross-sectional properties of both isotopes are primary determinants of overall polonium activity in the reactor. Therefore, understanding the difference between Bi-209 and Pb-208 in terms of capture cross sections would be helpful to explain the different polonium buildup behaviors. For this reason, capture cross sections of Bi-209 and Pb-208 are given in the same plot in Figure 4.8.
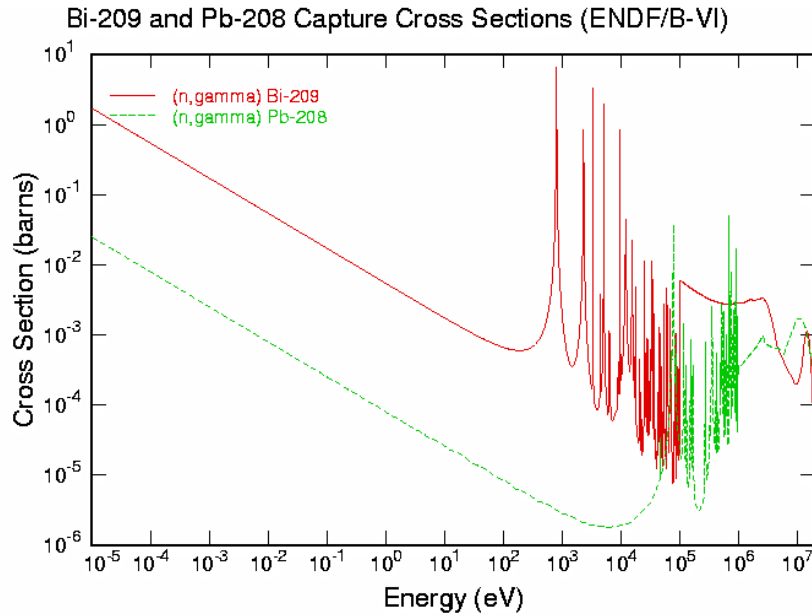
Figure 4.8   Bi-209 and Pb-208 Capture Cross Sections (ENDF/B-VI)[85]

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[85] Personal contact with Dr. Won Sik Yang of Argonne National Laboratory on August 4, 2006

As we see in Figure 4.8, the Pb-208 capture cross section is two orders of magnitude smaller than that of Bi-208. Hence, all things being equal, we expect to see much higher Po-210 activity results for LBE coolant.

Various institutions around the world have made rigorous calculations on Po-210 activation in LBE coolant. Specifically, there are two results I discuss in this study.

Argonne National laboratory has been leading the design efforts for liquid metal cooled reactors. I attended two day seminar in Argonne National Laboratory on August 3-4, 2006. Below is the discussion of the assessment Po-210 build up that was discussed at Argonne National Laboratory (ANL).

The polonium activity estimation was done under the following parameters:

- Core average flux is $5.40*10^{14} n/cm^2 s$.
- Core average one group capture cross sections of Bi-209:
  3.02 mb (ENDF/B-VI)
  5.28 mb (ENDF/B-V)
- Coolant volume fraction of active core is 0.278.
- Primary coolant volume is 64.8 m^3.

Figure 4.9 shows Po-210-specific activity in LBE coolant based on ANL calculations.
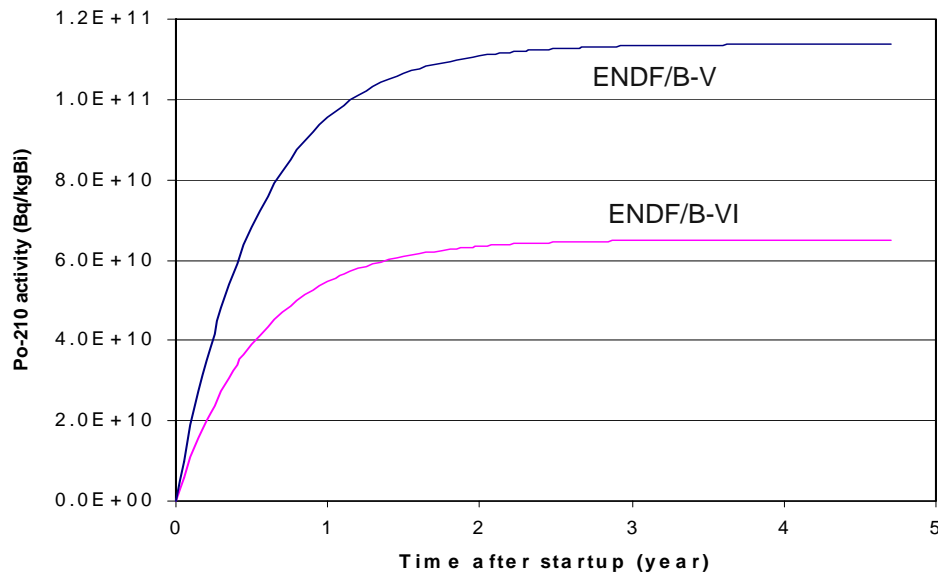
Figure 4.9   Po-210 Activity in LBE Coolant (Bq/Kg)

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As seen in the Figure 4.9, Po-210 buildup is calculated to reach over $6x10^{10}$ Bq/kg after two to three years of irradiation with the use of the ENDF/B-VI capture cross section library. However, if ENDF/B-V capture cross section library is used, Po-210 activity is an order of magnitude greater. Obviously, uncertainty in the cross sections is a part of problem in resulting in divergent activation calculations.

Another research institution I contacted to share my results was the ENEA (Italian National Agency for New Technologies Energy and Environment). The polonium activity estimation in this study was done under the following parameters:

- Core average flux is $1.19*10^{13} n/cm^2 s$. (averaged on the coolant volume)
- Core average one group capture cross sections of Bi-209 is 6.4 mb

After one year irradiation, with an average flux on the coolant of $1.19*10^{13} n/cm^2 s$, Po-210 activity reaches to $3.1*10^{17}$ Bq in 1936 tons of lead-bismuth. This makes $1.6*10^{11}$ Bq per kilogram. It is a factor of two to three times higher than Argonne's calculations. However, I need to emphasize significant differences in terms of bismuth cross section and flux levels used in the calculations.

The Po-210 activity in LBE coolant after five years of irradiation based on the calculations performed in this thesis is given in Figure 4.10.
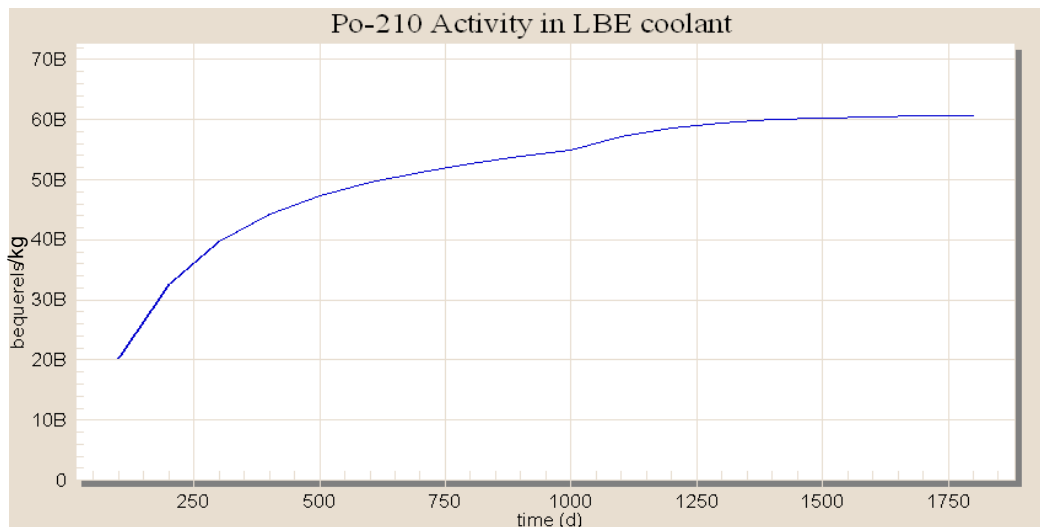
Figure 4.10   Pb-210 Activity in LBE Coolant (Bq/Kg)

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Table 4.1 provides a summary of results for calculations of LBE in this thesis. As seen in this table, the overall activity in LBE coolant is dominated by Po-210 and Bi-210. As far as the activity is concerned, Po-210 and Bi-210 contribute more than 90% of the total activity.

	1300 (Days)	1400 (Days)	1500 (Days)	1600 (Days)	1700 (Days)	1800 (Days)
Bi-210	6.176E+10	6.172E+10	6.169E+10	6.169E+10	6.171E+10	**6.175E+10**
Po-210	5.953E+10	6.004E+10	6.034E+10	6.052E+10	6.064E+10	**6.072E+10**
Bi-212	7.307E+07	9.050E+07	1.100E+08	1.314E+08	1.546E+08	1.793E+08
Bi-211	9.751E+04	1.017E+057	1.062E+05	1.108E+05	1.155E+05	1.202E+05
Pb-205	1.092E+05	1.183E+05	1.274E+05	1.365E+05	1.455E+05	1.546E+05
Bi-208	3.821E+04	4.157E+04	4.493E+04	4.829E+04	5.165E+04	5.501E+04
Bi-210m	3.392E+04	3.680E+04	3.968E+04	4.255E+04	4.543E+04	4.830E+04
Bi-213	1.394E+04	1.755E+04	2.169E+04	2.634E+04	3.151E+04	3.717E+04

Table 4.1   Selected Nuclides with High Activity Rates after 5 Years Irradiation (Bq/Kg)

Whereas the Po-210 activity in a lead system is about $8x10^6$ Bq/Kg, the polonium activity in LBE reaches $6x10^{10}$ Bq/Kg. There is a difference of almost four orders of magnitude. From a polonium activation standpoint, this is a major advantage of lead over LBE coolants. Further, the four orders of magnitude difference in coolant activation levels is consistent with the in the literature.[86]

What causes this difference is another interesting point to study further. Although this is not the main concern of this thesis, one may argue that capture cross sections of Bi-209 and Pb-208 are the primary factors affecting the prediction of polonium activity.

As we expect to see, due to decay, polonium buildup is not linear. Po-210 reaches a constant value after about 3-4 years, when the decay of Po-210 (half-life of 138 days) is approximately equal to the number production of Po-210 from Bi-209.

At this point, it is valuable to summarize the three different research results related to Po-210 activation in LBE coolant. Table 4.2 summarizes the three different calculations from ANL, ENEA and this thesis.

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[86]Kamil Tucek, "Comparison of SFRs and LFRs as Waste Burners", Nuclear Engineering and Design, 236 (2006), 1589-1598. They quote that pure lead produces less polonium than LBE by a factor of 10^4.

	Flux Level	Irradiation Time	Bi209 X-Section	Po210 Build up (Bq/Kg)	Code Used.
ANL	5.5E+14	5 Years	3.2 mb	6.8E+10(interpolated)	MCNP
ENEA	1.19E+13	1 Years	6.4 mb	1.6.E+11	MCNPX SP-FISPACT
Our Results	5.5E+14	5 Years	3.2 mb	6.072E+10	ORIGENARP

Table 4.2   Comparison of Different Calculations on Po210 Activity in LBE Coolant

Figure 4.11 and Figure 4.12, in linear and logarithmic scale, respectively, demonstrate exponential decay behavior based on the calculations in this thesis. Note that Po-210, with a relatively short half-life, undergoes rapid alpha decay.
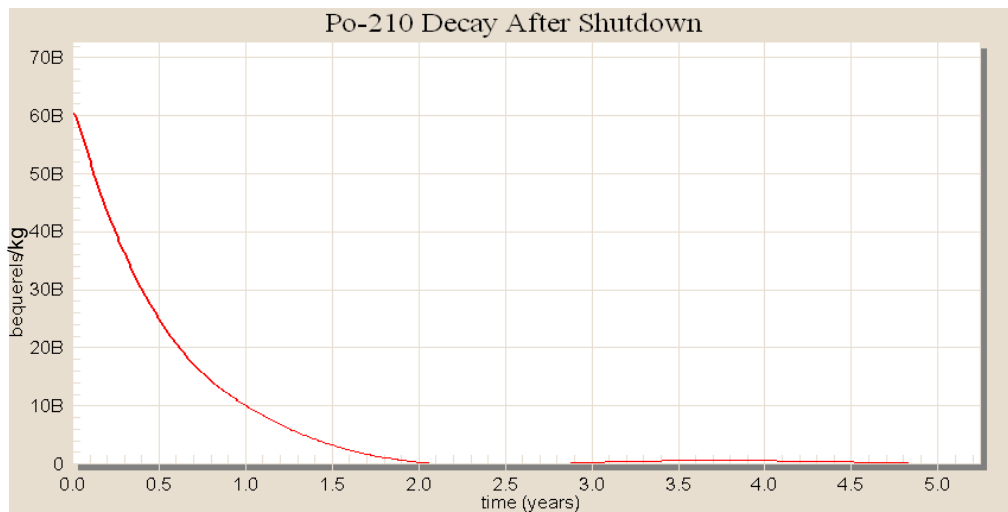
Figure 4.11   Po-210 Activity Decay after Shutdown in Linear Scale (Bq/Kg)
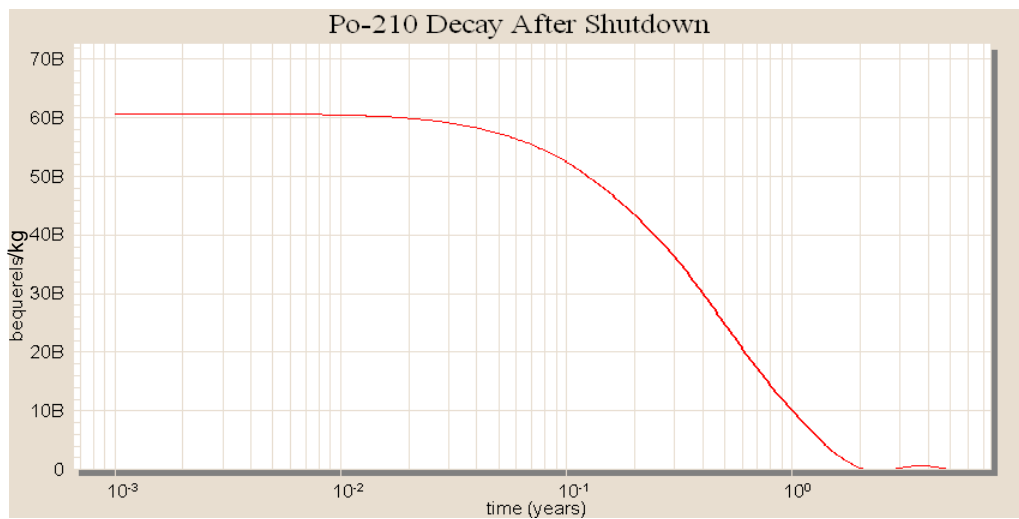
Figure 4.12   Po-210 Activity Decay after Shutdown in Log Scale (Bq/Kg)

Along with Po-210, the production of long-lived activation products in the LBE coolant is another potential concern. Long-term alpha activity of typical LBE coolant comes from Bi-210m with $T_{1/2} = 3.6*10^6\,year$. The radionuclide Bi-210m is generated by the reaction of Bi-209(n,γ) and decays with a half-life of $3.6*10^6\,years$.

Finally, the alpha active radionuclide Po-210 buildup is a primary concern in both lead- and LBE-cooled reactor systems. However, while polonium activity in lead is about $8x10^6\,Bq/Kg$, polonium activity in LBE reaches $6x10^{10}\,Bq/Kg$. This is a difference of almost four orders of magnitude. From a polonium activation standpoint, this is a major advantage of lead over LBE coolants

3. Sodium (Na)

The natural isotope of sodium is Na-23. It represents 100% of sodium in nature. Therefore, Na-23 was used as an initial isotope in the simulation. The process of neutron capture while sodium is flowing through the core, leads to the formation of the Na-24 isotope with a half-life of 15 hours.[87] In the reactor, Na-24 and Na-22 are generated as a result of the nuclear reaction:

$$Na^{23}(n,\gamma) \rightarrow Na^{24} \xrightarrow[15hours]{2n} Na^{22} \tag{4.8}$$

Based on the calculations in this thesis, the Na-24 activity in sodium coolant after five years of irradiation is given in Figure 4.13. Over the very first year of the irradiation, Na-24 builds up very quickly and the reaches equilibrium in two to three years.
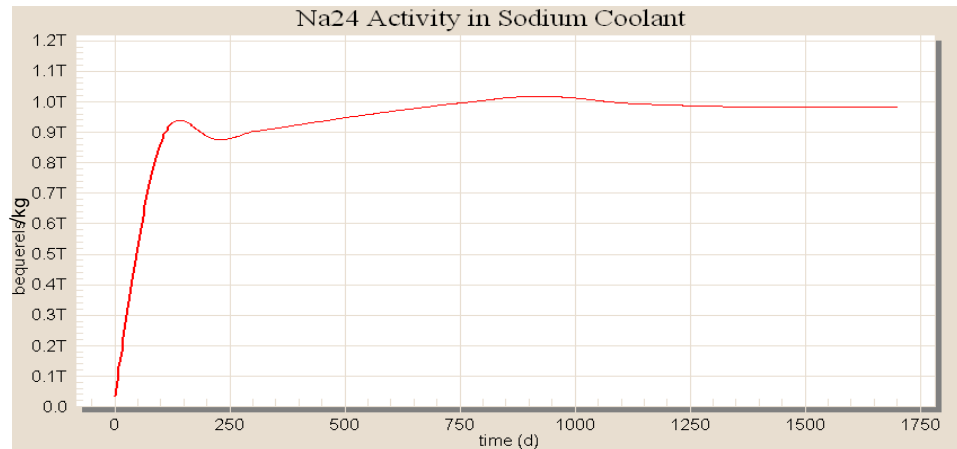
Figure 4.13   Na-24 Activity in Sodium Coolant (Bq/Kg)

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[87] IAEA-TECDOC-1289.

Na-24 activity, which gives rise to a requirement for protection against gamma radiation, reaches a level of nearly 10^{12} Bq/Kg of sodium.

Table 4.3 shows generation of activation products of sodium during five years of irradiation.

	1300 (Days)	1400 (Days)	1500 (Days)	1600 (Days)	1700 (Days)	1800 (Days)
Na24	9.896E+11	9.851E+11	9.826E+11	9.815E+11	9.814E+11	**9.820E+11**
Na22	1.453E+07	1.452E+07	1.452E+07	1.451E+04	1.451E+04	**1.451E+07**

Table 4.3   Selected Nuclides with High Activity Rates after 5 Years Irradiation (Bq/Kg)

As seen in Table 4.2, the major radioactivity in the sodium is due to Na-24. Calculations for the biologic protection of the reactor are based on this nuclide.[88] However, because its half-life is only 15 hours, it is of limited importance during extended reactor shutdown; in such cases, only nuclides with half-lives of at least several days are of interest.[89]

Although a sodium-cooled SSTAR type reactor has not yet been built, sodium technology has been well tested and studied over the years. Hence, it seems worthwhile to take a look at operational activity levels of some of the sodium-cooled fast reactors that were and have been in operation around the globe. The maximum Na-24 and Na-22 activity levels in some of the major sodium-cooled reactors are given in Table 4.4.[90]

It is interesting to note that reactor coolant system design and in particular, coolant residence time in the core, are factors affecting the equilibrium activity levels.

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[88] IAEA-TECDOC-687, "Fission and Corrosion Product Behavior in Liquid Metal Fast Breeder Reactor (LMFBRs)", February 1993.

[89] Ibid.

[90] Activity levels are taken from IAEA-TECDOC-687.

	Operating Country	Power	Na24 15h (TBq/Kg)	Na22 2.6a (MBq/Kg)	Still Operated
EBR-II	USA	62.5 MWth	**0.1**	**5**	Shut Down (1995)
FFTF	USA	400 MW th	**0.41**	**20**	Shut Down (1992)
PFR	UK	250 MWe	**-**	**14**	Shut Down (1994)
Rapsodie	France	40 MWe	**-**	**40**	Shut Down (1983)
Phenix	France	233 MWe	**-**	**22**	In Operation
BOR-60	Russia	60MWth	**2.5**	**22**	Shut Down
BN-350	Kazakhstan	150MMWe	**0.37**	**28**	Shut Down (1999)
BN-600	Kazakhstan	600MWe	**0.75**	**26**	In Operation

Table 4.4   Maximum Na-24 and Na-22 Activity Levels in Selected Sodium-Cooled Fast Reactors

Figure 4.15 shows the results of a simulation in this thesis that evaluated the decay of activity in sodium coolant following reactor shutdown.
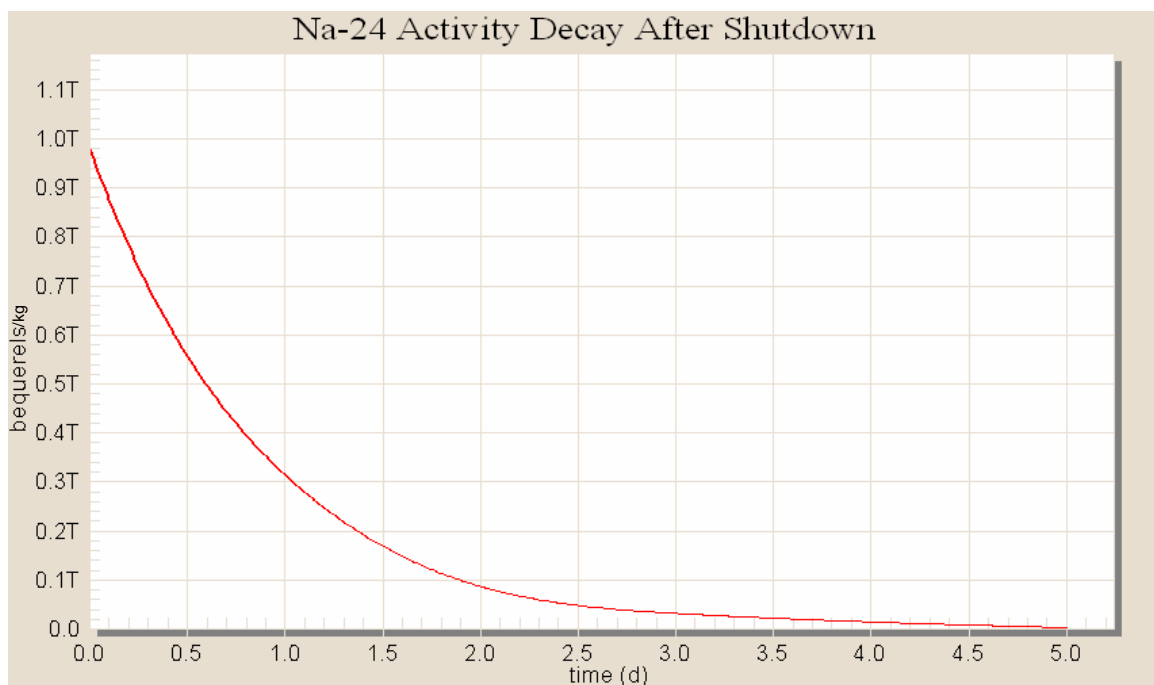
Figure 4.14   Total Activity Decay of Sodium in 5-Year Cooling Down Period

It follows from the figures that Na-24 activity decreases rapidly over a five-day cooling down period. Neutrons do not generate sodium activation products with half-lives of more than 2.6 years, even over the course of 50 years of exposure to an intense neutron

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flux of $10^{16} cm^{-2}s$.[91] After 50-100 years of cooling down, the used sodium coolant might be free enough of radioactivity to be used in industry or returned to nature.[92] This is a rather unusual result. There are only about fifteen to twenty chemical elements of the periodical table that could be exposed to such a flux, and would decay sufficiently during 50-100 years to allow such an exemption.[93]

4.   Tin (Sn)

Without doubt, tin is the least studied coolant among those I have selected. Hence, it is hard to find considerable literature about the activation characteristics of tin.

Tin is the element with greatest number of stable isotopes (ten). Moreover, 28 additional unstable isotopes are known. Naturally-occurring isotopes of tin and their natural abundances are as follows:

Isotopes and Their Natural Abundances (%)									
Sn112	Sn114	Sn115	Sn116	Sn117	Sn118	Sn119	Sn120	Sn122	124
0.97	0.65	0.34	14.54	7.68	24.23	8.59	32.59	4.63	5.79

Table 4.5   Naturally Occurring Isotopes of Tin

In the calculations of this thesis, tin was represented with naturally occurring distribution of isotopes. The results of the simulation in terms of total activity of tin coolant are given in Figure 4.15.
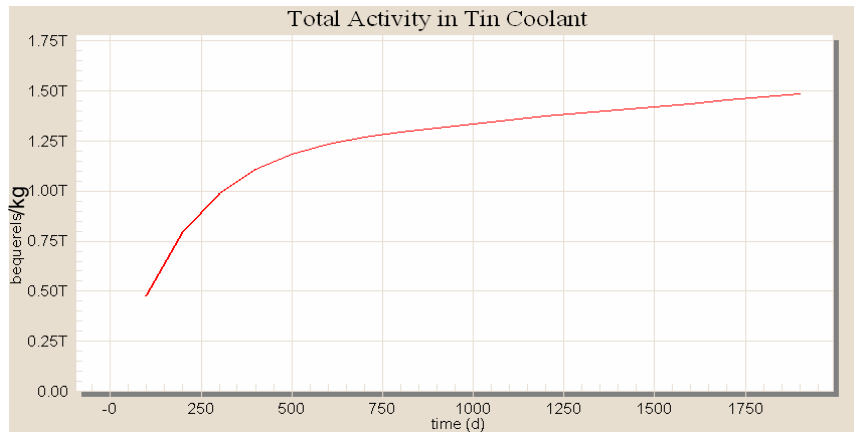
Figure 4.15   Total Activity of Tin Coolant (Bq/Kg)

[91] V.I Oussanov, D.V. Pankratov, E.P. Papov, P.I Makelov, L.D Riabaya, S.V Zabrodkaya, "Long-Lived Residual Activity Characteristics of Some Liquid Metal Coolants for Advanced Nuclear Energy Systems" 1999, IPPE, Russia.

[92] Ibid.

[93] Ibid.

More important than the numerical value of the activity, the complete absence of alpha active nuclides, such as Po-210 and Pb-210, makes tin less radiotoxic than lead and LBE.

F. SUMMARY OF THE ACTIVATION CALCULATIONS

All of the selected coolants: lead, LBE, sodium and tin, were irradiated with the same neutron flux under the same neutron spectrum characteristics of SSTAR. Activation products and their accumulations, particularly hazardous radionuclides, were compared with similar studies where available in order to verify the accuracy of our results.

The irradiation of lead and LBE coolants produce a substantial amount of Po-210 over time. Simulation results have shown that Po-210 activation in LBE coolant is of the same order of magnitude, 10^{10}, of the study performed by the Argonne National Laboratory,[94] but a factor of three less than the study prepared by ENEA, Italy.[95] There are a number of explanations for this discrepancy. First of all, different computer codes were used in these calculations. Whereas our calculations are based upon OrigenArp using Origen-S interface, the studies conducted by Argonne National Laboratory and ENEA are based upon Monte Carlo particle transport codes which allow more realistic simulation with fewer constraints. Another factor which leads to discrepancy would be the difference between the one-group cross-section of Bi209. In this study I used the ENDF/B-VI library. It is worthwhile to emphasize the fact that the one-group cross-section based on ENDF/B-VI is only 57% of that based on the ENDF/B-V data. More importantly, the flux levels, averaged on the coolant volume, differ significantly. These three factors might account for the discrepancy among the different calculations.

The accumulation of Po-210 was four orders of magnitude greater in the LBE coolant than in lead coolant. Although lead and lead-bismuth coolants produce other alpha activities, like Pb-210, and gamma active isotopes, such as Bi-207 and Bi-208, the production of polonium poses a particular hazard since, in case of an accident with

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[94] Study was presented by Won Sink Yang on August 3, 2006 in Argonne National Laboratory. Polonium activation build up is shown in Figure 4.9.

[95] "Evaluation of Spallation and Activation Products in Primary System" by ENEA. Document was prepared by ENEA, Italy and taken through personal contact with Dr.Carlo Petrovich. In the report, Po210 activity for 20 year irradiation was calculated as 3.71E17 Bq/Kg for 1936 tons of LBE.

rupture of the reactor vessel and a release of nuclides into the atmosphere, its radiotoxicity is equivalent to the harmful effects of all radionuclides put together.[96]

Activation calculations for sodium carried out in this study are parallel to operational experience gained at different sodium-cooled fast reactors, as shown in Table 4.3. Sodium's activation behavior is more favorable than both lead and LBE. Simulation results showed that the primary activation product in sodium coolant after five years of irradiation is Na-24 with a 15-hour half-life. Sodium activation chains accumulated no radionuclide with a half-life longer than 2.6 years.

Activation calculations for the tin coolant were also carried out, although tin has yet to be of primary interest to scientists. There is little or no information about coolant activation products in the literature. Tin activation calculations presented here demonstrate that the activated tin coolant would be free from hazardous alpha and gamma radionuclides.

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[96] G.L. Khorasanov; A.I. Blokhin, "Fundamentals for the Development of a Low-Activation Lead Coolant with Isotopic Enrichment for Advanced Nuclear Power Facilities", 2001.

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V. CONCLUSION

No single property of the candidate coolants is able to uniquely define the outcome of the coolant selection process for ongoing efforts to build small reactors in the coming decades. When considering coolant options, there have always been a set of advantages and disadvantages, which are evaluated and taken into consideration in this process. However, sometimes available technology, natural resources, or even political motivations might be part of the equation in the selection process.

So far, the physical, thermohydraulic properties, and neutronic characteristics of lead, LBE, sodium, and tin coolants have been discussed in great detail. The intent of this chapter is two-fold. The first objective is to summarize the favorable and unfavorable properties of coolants in tabular form. The second is to offer recommendations for future thesis studies on reactor coolants.

In this thesis, I chose not to rank the coolants according to the number of favorable properties, since that sort of evaluation would require application of weighing factors and assigning values according to their importance. However, there are some significant advantages and disadvantages of selected coolants, which need to be emphasized further.

Po-210 build up is a serious concern for both lead and LBE coolants. Giving the fact that Po-210 build up is four orders of magnitude higher in LBE than in lead, the choice between lead and LBE may be dominated by this factor. On the other hand, although sodium has nearly excellent thermophysical properties, its chemical reactivity with water and air poses serious drawback. Like sodium and LBE, tin has a serious shortcoming. Almost no possible structural material is compatible with tin due its corrosive behavior.

Finally, the advantages and disadvantages of selected heavy metal coolants are presented in tabular form.

A. LEAD

The advantages and disadvantages of lead as a coolant are presented in Table 5.1. They are categorized as physical and thermal properties, neutronic/radioactive properties, and other properties, including abundance, cost, etc.

	Advantages	Disadvantages
Physical and Thermal Properties	• Does not react with water and air violently. • Lead has a high boiling point (1740^0C), avoiding a coolant boiling problem. • Favorable heat transport properties. • Offers the potential for natural circulation, which may lead to a new level of plant simplification and inherent safety.	• Lead has a relatively high melting point (327.4^0C), which limits the operational temperature level of the reactor. • Redundant heaters may be required to avoid problems with freezing and blockages. • Density is 10 times higher than sodium, causing some drawbacks in the overall system. • Lead is highly corrosive with many possible reactor structural materials. Precautions need to be taken for implication.
Neutronic/ Radioactive Properties	• Energy loss due to elastic scattering is significantly smaller than sodium. [97] • Higher scattering without increasing moderation for neutrons prevents escaping and provides an excellent reflecting capability.	• Primarily Po-210 and other alpha active radioactive nuclides accumulate after 5-year irradiation. • Residual activity of coolant is high due to Pb-205, half-life $1.5 \ 10^7$ years.[98] • Problems with radioactive waste management and coolant disposal during decommissioning.[99]
Other	• Lead is abundantly available in nature. • Relatively low cost on world market ($1/Kg).	• Application of lead coolant in reactor systems will require further studies on material compatibility and isotropic tailoring to reduce polonium buildup.

Table 5.1   Advantages\Disadvantages of Lead Coolant

B. LEAD-BISMUTH EUTECTIC COOLANT (LBE)

LBE coolant has similar physical and neutronic characteristics to lead. However, the introduction of bismuth brings both favorable and unfavorable aspects to the eutectic coolant. Most importantly, as discussed in Chapter IV, polonium buildup increases by four to five orders of magnitude due to presence of the Bi-209 isotope. Table 5.2 shows the advantages and disadvantages of (LBE) coolant.

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[97] Kamil Tucek; Johan Carlsson; Hartmut Wider, "Comparison of Sodium And Lead-Cooled Fast Reactors Regarding Severe Safety And Economical Issues", 13th International Conference on Nuclear Engineering, Beijing, China, May 16-20, 2005, ICONE13-50397.

[98] IAEA-TECDOC-1289, p.63.

[99] Ibid.

	Advantages	Disadvantages
Physical and Thermal Properties	- Does not react with water and air violently. - LBE has a high boiling point (1670^0C), avoiding a coolant boiling problem. - LBE has favorable heat transport properties. - Offers potential for natural circulation, which may lead to a new level of plant simplification and inherent safety. - LBE has a relatively low melting point (123.5^0C) compared to lead, limiting problems with freezing in the system.	- High density causes some drawbacks in overall system. - LBE is highly corrosive with many possible reactor structural materials. Precautions need to be taken for this implication. - LBE requires high pumping power. Pumping power requirement for LBE is 6-7 times greater than for sodium.
Neutronic/ Radioactive Properties	- Energy loss due to elastic scattering is significantly smaller than sodium.[100] - Higher scattering without increasing moderation for neutrons prevents escaping and provides an excellent reflecting capability. - Neutron economy of LBE coolant is better than sodium.[101]	- Primarily Po-210 and other alpha active radioactive nuclides accumulate after 5-year irradiation. - Po-210 accumulation in LBE coolant four orders of magnitude greater than lead coolant. - Residual activity of coolant is high due to Bi-210, half-life $3.6 \ 10^6$years.[102] - Problems with radioactive waste management and coolant disposal during decommissioning. [103]
Other	Technology can be considered mature based on the Russian experience.	- LBE coolant technology has been used on Russian submarines but it has not been tested on a commercial nuclear power plant - Bismuth is a relatively scarce and costly material, the capital cost of LBE increases with the fraction of bismuth used. - Application of LBE coolant in reactor systems will require further studies on material compatibility and isotropic tailoring to reduce polonium buildup.

Table 5.2   Advantages\Disadvantages of LBE Coolant

C.   SODIUM

Sodium offers superior physical and thermal properties and does not have most of the shortcomings of lead, LBE, and tin coolant. Therefore, it has been used by a number of nations as a reactor coolant for more than five decades. While several power reactors

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[100] Kamil Tucek; Johan Carlsson; Hartmut Wider, "Comparison of Sodium And Lead-Cooled Fast Reactors Regarding Severe Safety and Economical Issues", 13th International Conference on Nuclear Engineering, Beijing, China, May 16-20, 2005, ICONE13-50397.

[101] Ibid.

[102] IAEA-TECDOC-1289, page 63.

[103] Ibid.

have been shut down, BOR-60, JOYO, Phénix, and BN-600 are still operating—the latter being in quasi-commercial operation since 1982.[104] New sodium-cooled reactors are under construction in Russia, China, and India.[105] Table 5.3 shows the advantages\disadvantages of sodium coolant.

	Advantages	Disadvantages
Physical and Thermal Properties	• Superior heat transport properties compared to lead, LBE, and tin. • Sodium shows good compatibility with many metals even at high temperature. • Low density allows passive safety rod operation (by gravity force). • Due to low density, pumping power required is less than lead, LBE, and tin. • Due to its low melting and boiling point, sodium coolant requires the lowest fuel cladding temperature.	• Sodium violently reacts with water and air. • Melting temperature is 98^{0}C, so heaters are required to maintain the liquid phase of coolant. • A low boiling point, on the other hand, raises safety concerns for coolant boiling during operations.
Neutronic/ Radioactive Properties	• No alpha active particle accumulation, such as Po-210. • Neutrons do not generate in sodium radionuclides with a half-life of more than 2.6 years.	• Leakage of neutrons and their contribution to overall neutron balance in the system is more significant for sodium.[106] • Gamma active nuclide Na-24 accumulates in sodium after irradiation.
Other	• Over the years, there has been extensive operational experience with sodium reactors.	• Loss of coolant increases reactivity, positive reactivity voids coefficient.

Table 5.3 Advantages\Disadvantages of Sodium Coolant

D.   TIN

One of the first proposals to use liquid tin for a nuclear power facility was done by J. R. Weeks in 1971. He considered melts of natural tin and its eutectic alloys, Bi-Sn, Pb-Sn, and Bi-Pb-Sn, as potential coolants for fast reactors. The attractive features of tin, such as a high boiling point (2270°C), low vapor pressure (~100 Pa for 800°C), and low

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[104] Kamil Tucek; Johan Carlsson; Hartmut Wider, "Comparison of Sodium And Lead-Cooled Fast Reactors Regarding Severe Safety and Economical Issues", 13th International Conference on Nuclear Engineering, Beijing, China, May 16-20, 2005, ICONE13-50397.

[105] Ibid.

[106] Ibid.

melting point (231.9 °C), give tin some advantages over lead and LBE. Table 5.4 shows the advantages\disadvantages of tin coolant.

		Advantages	Disadvantages
Physical and Thermal Properties		• Tin has a high boiling point (2270^0C), avoiding coolant boiling and providing a wider range of operating temperatures. • Some thermal and physical properties (heat conductivity, heat capacity, dynamic viscosity) are better than LBE and lead. • The low melting point of tin in comparison with lead allows the inlet temperature of a coolant to decrease. • Due to tin's thermal and physical properties, very effective heat removal, up to 1 GW/m3, is possible. In this case, the required coolant temperature is on the order of 400-600°C and coolant flow velocity is about 2-4 m/s.[107]	• Highly corrosive with many potential structural materials. • Tin has a relatively high melting point (232^0C), which limits the operational temperature level of the reactor. • Redundant heaters may be required to avoid problems with freezing and blockages.
Neutronic/ Radioactive Properties		• The complete absence of alpha and gamma active nuclides (Po-210, Pb-210, and Na-24) in the radioactive wastes of tin makes it less radiotoxic than lead and LBE.	
Other			• Tin is a relatively costly material ($2.5/Kg). • It has not been thoroughly studied and tested thus far.

Table 5.4   Advantages\Disadvantages of Tin Coolant

E.   RECOMMENDATIONS

As predicted by the Special Report on Emission Scenarios (SRES) initiated by the Intergovernmental Panel on Climate Change (IPCC), the energy demand of primary sources will increase between 1.7- and 3.7-fold until 2050 (INPRO, 2003). The share of nuclear power will increase considerably by 2050, with a median of more than 4 times.

Small, secure, transportable reactors will provide a reliable, safe, proliferation resistant option both for developing nations in the coming decade. The optimal choice of core, structural, and coolant materials for SSTAR systems are still an open question.

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[107] Bruce W. Spencer, "The Rush to Heavy Liquid Metal Reactor Coolants Gimmick Or Reasoned", ICONE-8729, April 2-6, 2000, Baltimore, MD USA.

However, the requirement of a fast neutron spectrum for efficient breeding and TRU incineration implies the usage of coolants with low moderating power, such as sodium, lead, LBE, or tin.

In this thesis, two types of liquid metals, lead and sodium, proposed in GEN-IV systems, were studied in addition to LBE and tin. Their physical and thermal properties were explored and the activation products accumulated in the coolants due to irradiation were calculated by using the analytical tool, OrigenARP.

Recommendations for future thesis work in this area are made as follows:

- OrigenARP analytical tool was used in this thesis. OrigenARP is an automated sequence to perform isotopic depletion/decay calculations using the ARP and ORIGEN-S codes of the SCALE system. For future activation studies, other computational tools, such as MCNP (Monte Carlo N-Particle) and ALARA[108] (Analytic and Laplacian Radioactivity Analysis) codes can be used to compare results obtained in this thesis.

- Bi-209 cross sections in different nuclear data differ significantly from each other. One group cross section based on ENDF/B-VI is only 57% of that based on ENDF/B-V data. Uncertainty about Bi-209 can be seen in Figure 5.1. Therefore, a reevaluation of lead and bismuth cross sections might be necessary for a more accurate evaluation of lead alloy coolant activation.

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[108] A new activation code released in January 1998 and developed specifically for analysis of radioactivity in fusion energy systems.
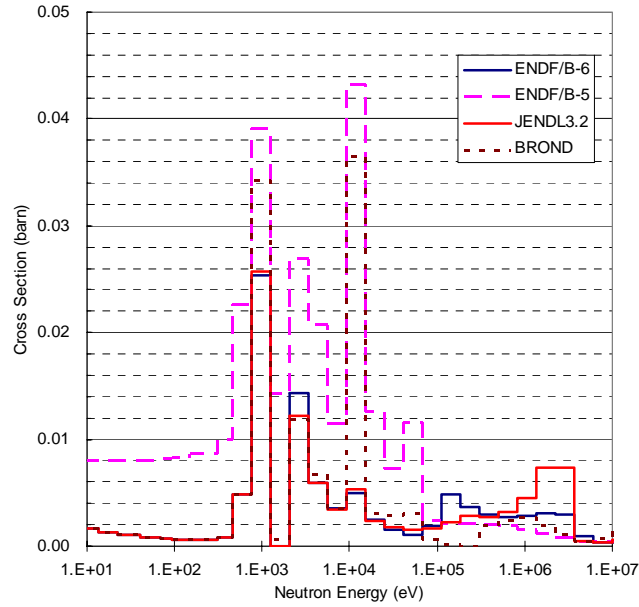
Figure 5.1 Bismuth Cross Sections in Different Libraries[109]

- One of the findings of this thesis is that little is known about tin as a reactor coolant. Although some thermal properties are favorable for heat transfer in reactor systems, it has never been considered a candidate due to its corrosive behavior and compatibility problems with structural material. It seems that there are no plans in the foreseeable future to build tin- or tin alloy-cooled reactors in the coming years. Hence, tin as a reactor coolant should not considered in future studies.

- Finally, when considering the array of selected candidate coolants, I conclude that sodium and lead have the most promise for future application.

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[109] From Dr. Won Sink Yang's presentation on Polonium Activation in LBE Cooled Reactors Argonne National Laboratory, August 3,4 2006.

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